

# 2019 NE-4 Nuclear Fuel Cycle and Supply Chain (NFCSC) Achievements Report

January 2020

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Prepared for the
U.S. Department of Energy
Office of Nuclear Fuel Cycle and Supply Chain
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

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Compiled and edited by:
Robert Rova (Robert.Rova@nuclear.energy.gov)
Bonnie Hong (bonnie.hong@inl.gov)
Katie Stokes (katherine.stokes@inl.gov)

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# 2019 NE-4 Nuclear Fuel Cycle and Supply Chain (NFCSC) Achievements Report

#### 1. INTRODUCTION

The United States Department of Energy, Office of Nuclear Energy (DOE-NE) in 2019 established new and re-established existing presidential and departmental nuclear energy priorities:

- Support the Administration's efforts to achieve U.S. energy dominance and economic prosperity through technology leadership
- Support DOE-NE mission priorities and goals for the existing nuclear fleet, advanced reactor pipeline, and national fuel cycle infrastructure
- Focus on early stage research, and effective cooperation with industry, academia, and other government agencies
- Propose investments to maintain and improve capabilities and infrastructure to conduct leading-edge science, research, and technology development.

DOE-NE mission priorities are focused on the existing nuclear fleet, advanced reactor pipeline, and fuel cycle infrastructure. Therefore, the DOE office of Nuclear Fuel Cycle and Supply Chain (NFCSC) NE-4 Program will align research and development (R&D) efforts to (1) support innovative reactor technologies that may offer improved safety, functionality, and affordability, and (2) conduct research to reduce long-term technical barriers for advanced nuclear energy systems (NES). Key activities to execute this mission include:

- R&D planning to support the building of a versatile advanced test reactor by 2026
- R&D of advanced nuclear fuels covering the entire spectrum of existing and advanced NES
- Analyze specific NES to identify benefits and challenges, leading to the development of common technology R&D goals
- Contribute to a broader understanding of the required characteristics of NES that can provide substantial improvements to the current once-through fuel cycle option in the U.S.
- R&D of advanced Material Recovery and Waste Form technologies that improve current fuel cycle performance through minimizing processing, waste generation, and potential for material diversion
- Assessment of security vulnerabilities of advanced fuel cycles and development of management and safeguards technologies and systems to address the risks.

To achieve the NE-4 mission priorities, the DOE Office of NFCSC supports two programs. The Fuel Cycle Research and Development program involves 10 major R&D campaigns to engage in impactful research, development, demonstration, and deployment (RD&D). DOE created the Versatile Test Reactor (VTR) program in April 2018.

#### Fuel Cycle Research and Development (FCRD) Program

The Systems Analysis and Integration (SA&I) Campaign provides an integrated view of nuclear energy systems by performing analyses and studies, and interacts with all NFCSC campaigns.

- The Advanced Fuels Campaign (AFC) performs RD&D activities to identify and develop innovative fuels, cladding materials, and other associated technologies to improve the performance and safety of current and future reactors. The AFC also increases the efficient utilization of nuclear energy resources, contributes to enhancing proliferation resistance of the nuclear fuel cycle, and addresses challenges related to waste management and ultimate disposal issues.
- Joint Fuel Cycle Studies (JFCS) is a collaboration with the Republic of Korea to assess the feasibility and nonproliferation of Electrochemical recycling to manage used fuel.
- The Material Recovery and Waste Form Development (MRWFD) Campaign matures advanced separation and waste processing technologies to support the various fuel cycle options defined in the DOE Nuclear Energy Research and Development Roadmap, Report to Congress, April 2010.
- The Material Protection, Control, and Accountability Technologies (MPACT) program develops innovative technologies and analysis tools to enable next-generation nuclear materials management for existing and future U.S. nuclear fuel cycles.

#### **VTR Program**

 The Versatile Test Reactor (VTR) program is one of the DOE-NE's highest priorities, and the VTR subprogram serves as a cornerstone to the Administration's focus on reviving and expanding the nuclear sector in the United States. Specifically, the VTR will support the modernization of United States infrastructure for early stage R&D.

This new research reactor will be capable of performing irradiation testing at much higher neutron energy fluxes than what is currently available today. This capability will help accelerate the testing of advanced nuclear fuels, materials, instrumentation, and sensors. It will also allow DOE to modernize its essential nuclear energy research and development infrastructure, and conduct crucial advanced technology and materials testing necessary to re-energize the U.S. nuclear energy industry.

# Fuel Cycle Research and Development Program

#### 2. SYSTEMS ANALYSIS AND INTEGRATION CAMPAIGN

Temitope Taiwo, Argonne National Laboratory (ANL), National Technical Director (NTD)

Brent Dixon, Idaho National Laboratory (INL), Deputy NTD

#### 2.1 Overview

#### 2.1.1 Mission

The SA&I Campaign provides a comprehensive view of nuclear energy system potential by performing analyses and studies and interacting with all NFCSC campaigns to support strategic planning for DOE-NE-4, allowing a refined focus on R&D activities. This step helps determine the technical and economic feasibility of advanced nuclear energy systems to inform DOE-NE-4 program development, planning, and budget formulation. Coordination of R&D contributes to the integration of DOE-NE-4 activities by identifying technology maturation needs and pathways, and informing decisions on infrastructure needs in a systematic manner.

This mission also supports national energy security needs for a diversified energy portfolio, which includes nuclear power for the long term.

#### 2.1.2 Campaign Objectives

The campaign has the following primary objectives:

- Perform high-quality analysis of nuclear energy systems to determine technical and economic viability, and identify benefits and challenges to: (a) provide insights on legacy and newly proposed reactors and nuclear energy systems; (b) develop an understanding of the role of nuclear energy and its competitiveness in current and future domestic and global energy markets; and (c) identify and assess strategies to improve economics, technical, and social sustainability of nuclear energy.
- 2. Develop tools, models, and processes to maintain our leading-edge capability in the world and ensure top-notch analyses of steady-state and dynamic behaviors of NES, including quantifying the impact of performance improvements, and studying issues associated with transition and deployment.
- 3. Facilitate integration of DOE-NE-4/NE R&D portfolio and strategy by working with NTDs and federal managers and office Directors, including assessment of programmatic risks of ongoing R&D by facilitating application of the Technology and System Readiness Assessment (TSRA) process by the R&D campaigns.

In 2019, the campaign activities were organized into three general areas, namely campaign management, Equilibrium System Performance (ESP), and Development, Deployment, and Implementation Issues (DDII), consistent with the SA&I Campaign objectives listed above (also, see Figure 1).

The campaign management area provides national technical leadership for the SA&I Campaign activities by: (1) planning and managing the activities in the campaign, (2) facilitating integration of R&D activities with the other DOE NFCSC program campaigns, and (3) contributing to integration of the Office of Nuclear Energy R&D activities in general, and that of the NFCSC program in particular, through systems analysis activities. The technical analyses within the SA&I Campaign are conducted under the ESP and

DDII work areas. The features and performance of alternative end-state NES are studied in the ESP work area. The issues and challenges of transitioning to and deploying the end-state systems, along with the competitiveness of nuclear in the future energy mix are studied in the DDII work area.

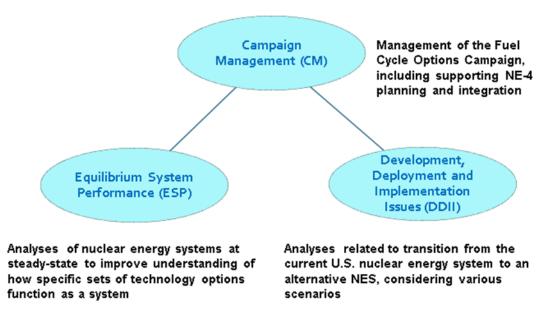


Figure 1. SA&I Campaign organization by work areas in 2019.

#### 2.1.3 Key 2019 Deliverables

Key deliverables for the campaign summarized results in each of the two technical areas. In 2019, the key deliverables/reports are:

#### **Equilibrium System Performance (ESP) Deliverables**

Report on the Algorithm for the Capital Cost Estimation of Reactor Technologies

- (ACCERT) Cost Algorithms Tool (June 2019)
- 2019 Analysis of Advanced Fuel Cycle Systems (March 2019)
- Daily Market Analysis Capability and Results (April 2019)
- Cost Drivers for Construction of DOE Large-Scale Nuclear Facilities (August 2019)
- Technical Insights on Newly Proposed Nuclear Energy Systems (September 2019)
- Office of Nuclear Energy Contributions to Beyond Levelized Cost of Electricity LCOE Phase 1 (September 2019).

#### **Development Deployment and Implementation Issues (DDII) Deliverables**

- Reassessing Methods to Close the Nuclear Fuel Cycle (March 2019)
- Technology Maturation Planning and Technology Roadmap Development Example Using the TSRA process (June 2019)

- Load Leveling Through Battery Electric Vehicles and Implications for Electric Power Generation (July 2019)
- 2019 Advanced Fuel Cycle Transition Analysis (August 2019)
- Application of CYCLUS to a Transition Scenario (August 2019)
- Adaptation of Oak Ridge Siting Analysis for Power Generation Expansion (OR-SAGE) for Advanced Nuclear Energy Systems (September 2019).

The following sections provide representative highlights of the SA&I Campaign work completed during the year.

### 2.2 Equilibrium System Performance

In the *Equilibrium System Performance* work area, the campaign performs analyses of nuclear energy systems to improve the understanding of how specific sets of technology options function as a system. This includes working with other campaigns to consider the effects of a range of specific implementing technologies for all parts of the nuclear energy system. It also includes the enhancement of the tools and capabilities for such analyses (e.g., for nuclear facility costing, economic, and competitiveness analyses), and development and maintenance of analysis data and libraries.

#### 2.2.1 Technical Insights on Newly Proposed Nuclear Energy Systems

The SA&I Campaign has been compiling information on the advanced nuclear energy systems currently under development by industries, universities, national laboratories, and foreign entities. In 2018, a compendium report was developed for informing on the fuel cycle performance of these nuclear energy systems. The content of the 2018 report was updated in 2019 to include information on 17 additional concepts. The set of evaluated nuclear energy systems now includes 73 advanced reactor concepts, 15 nuclear fuel concepts, and four reprocessing technologies. Nine national fuel cycle concepts proposed by foreign entities are also included (see Figure 2). Some general observations:

- The number of advanced reactor concepts has been increasing since 2000 and has accelerated recently in the United States because of investment of private capital and various initiatives of government organizations (DOE, NASA, Department of Defense). More than 75% of the advanced reactor concepts reviewed are small or microreactors targeting affordable construction cost and small grid markets in remote or isolated locations. High thermal efficiency, enhanced passive safety features, high security, and lower environmental impacts are commonly claimed for most of the advanced reactors.
- Traditional assumptions of coolant materials for thermal and fast neutron spectrum reactors have been
  changed in some advanced reactor concepts with use of coolants such as gas, molten salt, and lead, for
  example, considered for both thermal and fast spectra. Non-conventional cooling systems/
  technologies, such as heat pipe cooling and organic coolant, have been proposed.
- Currently, a number of the advanced reactor concepts are under design certification review or have been in contact with the U.S. Nuclear Regulatory Commission (NRC) or Canada Nuclear Safety Commissions for pre-application or vendor design review.
- Fuel cycles utilizing advanced reactor concepts are mostly in the Evaluation Group EG01 (current nuclear fleet based on once-through fuel cycle using enriched U fuel in thermal reactors) and EG02

(once-through fuel cycle using higher enriched U fuel to high burnup in thermal or fast reactors) of the 2014 Evaluation and Screening study, indicating they are targeting improvement of the fuel cycle performance of the current nuclear fleet. The next major collection of fuel cycles utilizing advanced reactor concepts are in EG23 or EG24 (continuous recycle of U, Pu, or TRU with natural uranium feed in fast reactors), which indicate some proponents/developers are aiming at improving sustainability of nuclear energy via a continuous recycle of U, Pu, or TRU using fast reactors. This trend is consistent with the conclusions of the evaluation and support study (i.e., the EG23 and EG24 are among the most promising fuel cycles).

• Except for external driven systems and evolutionary light water reactors (LWRs), advanced reactor concepts require High-Assay Low-Enriched Uranium (5–20%) as fissile material for igniting start-up cores or to compensate for poor neutron economy in small and microreactors.

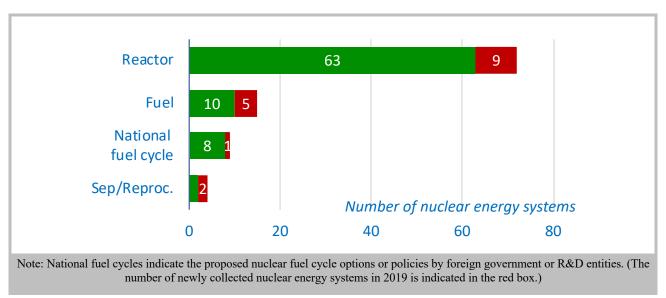


Figure 2. Tally of Nuclear Energy Systems Reviewed by SA&I Campaign.

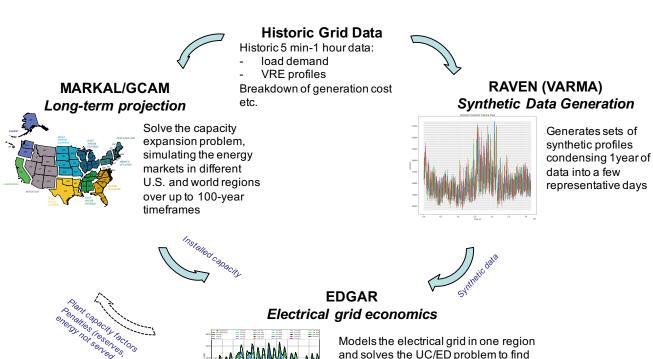
#### 2.2.2 Energy Market Economic Analysis Capability

Nuclear energy plays an important role in the U.S. energy mix and would need to be maintained or strengthened for the country to achieve significant greenhouse gas reduction. However, maintaining the nuclear portfolio is becoming increasingly challenging in the current U.S. energy market as the low price of natural gas and the penetration of subsidized and low-marginal cost variable renewable electricity are affecting the profitability of nuclear units. In this context, new nuclear power plants will need reduced capital and operations and management costs, enabling increased revenues by changes in market policies or increased flexible operation.

In 2019, the SA&I Campaign started a new focus on short-term electricity market analysis that included tools for power unit dispatch and nuclear deployment, all within broader macroeconomics and policy contexts. This allows conducting analyses to quantify the deployment and dispatch of nuclear energy in the

context of the modern complex electricity markets. The methods developed and codes acquired are displayed in Figure 3.

Capacity expansion problems are solved with the Global Change Assessment Model (GCAM) and/or MARKet and Allocation (MARKAL) models, simulating the energy markets in different U.S. and world regions to provide a scenario-based long-term perspective. The EDGAR (Economic Dispatch Genetic AlgoRithms) code solves the combined Unit Commitment and Economic Dispatch problems to find the optimal schedule of a fleet of generating units to meet the forecasted grid demand over the next day in deregulated markets with an hourly time resolution. The EDGAR code relies on sets of load demand and renewable generation data with a one-hour time-step generated out of historical data using the VARMA (Vector Auto-Regressive Moving Average) model in RAVEN (Risk Analysis Virtual Environment) to condense the full year of data into a few representative days. This full suite of market economic analysis codes was used to model the New York Independent System Operator region in order to demonstrate the capabilities acquired within the campaign. The NY-ISO region was selected because it is a mostly deregulated market with a significant fraction of nuclear and wind generations foreseen in 2050. This exercise was especially useful in order to understand the specificities of the different market modeling codes acquired and developed—the assumptions these codes rely on—and the remaining gaps in our tools. Daily market analyses were performed with EDGAR for a 40-unit model of NY-ISO operating in 2015 to demonstrate convergence of the obtained results and their sensitivity to different sets of synthetic data generated with VARMA. A similar analysis was performed for a long-term (2050) scenario produced by GCAM to confirm the installed capacity is sufficient to meet electricity demand, that reserve requirements are met every time of the year, and that the variable renewable energy (VRE) generation would not lead to excess generation if their curtailment is allowed.



Models the electrical grid in one region and solves the UC/ED problem to find the optimal fleet schedule that minimize electricity generation cost in deregulated





#### 2.2.3 Cost Drivers for Construction of DOE Large-Scale Nuclear Facilities

Successful facility construction projects tend to share certain characteristics that contributed to the successful outcome. An assessment was done in 2019 to identify those characteristics, and to develop a set of best practices and recommendations for the successful management of nuclear megaprojects, as funded by the DOE and implemented at U.S. national laboratories. ("Megaproject" is a literature technical term for a large, capital-intensive construction project, usually costing over \$1B.) The conclusions were derived from case studies of a group of four such projects or programs and a review of relevant project management literature. Contemporary interviews, news reports, and congressional debate transcripts supplement the factual research with information about how events were perceived at the time. From this information, issues and their root and contributing causes were identified. The projects or programs selected for the case studies were primarily the Spallation Neutron Source (SNS) and the Advanced Photon Source (APS). The Superconducting Super Collider (SSC) and the National Synchrotron Light Source-II were also selected and analyzed to gather additional insights, and to compare with SNS and APS. This total set of projects was selected to represent a suite of diverse facility types, management structures, and project outcomes. The APS and National Synchrotron Light Source-II projects were very successful in terms of schedule and budget execution; the SNS overcame a rocky start to achieve a largely successful result; and the SSC was cancelled after \$1.6B of capital had already been invested and substantial construction work had been completed. Each of these projects provides an interesting and informative lens into management processes and structures that can make a DOE large-scale nuclear project successful.

The literature review encompasses the substantial body of extant work on project management, and especially the sub-field of megaproject management. The case studies for each project were developed by assembling a detailed timeline of project events, external events (political and scientific), and conceptual and technical information that led to various events and decisions. Evolution in project cost and schedule estimates were tracked in as much detail as was available. Based on examination of relevant literature and the four case studies, conclusions were distilled in five categories: (1) overall project organization; (2) industry partnerships; (3) stakeholder engagement; (4) personnel recruiting and retention; and (5) human factors.

#### 2.2.4 Update of Fuel Cycle Cost Algorithms (ACCERT)

In 2019, work continued on the development of cost algorithms for improved estimation of the construction cost of conventional and advanced reactor concepts. The overall objective of the work was to increase the fidelity of the ACCERT algorithms, and consequently the usefulness and credibility of the evaluations performed with this tool. The following were accomplished in 2019:

Cost models of all the Nuclear Steam Supply System (NSSS)-supplied components were developed, for which costs were not provided in the reference cost databases (EEDB 1987). This work was initiated and substantially advanced in 2018, with the development of detailed cost models for the eight largest and most expensive parts of the NSSS. The work in 2019 completed that task, with an inclusion of all the remaining NSSS-supplied components that were not analyzed earlier. These include relatively minor, but numerous components, including rotating machinery, heat exchangers, demineralizers, tanks, etc., for several important functions of nuclear power plants. The reasonableness of the NSSS cost models was verified to give confidence in the cost models developed for the NSSS systems, at least at an aggregated level. This work offers, for the first time in the public domain, a complete set of models to evaluate the NSSS cost of advanced reactors systems.

- The number of cost models was substantially extended in order to include all the costs that contribute at least 0.5% of the total direct costs of the reference PWR12-BE. In order to accomplish this, a total of 29 cost models were newly developed in 2019. These, combined with the 31 cost models developed in the previous 2 years, reach a total of 60 cost models, with a cumulative contribution to the total direct cost of the reference PWR12-BE of more than 95%. With less than 5% of the direct cost of the reference design not directly quantified, the estimates performed with ACCERT now have a high degree of robustness.
- New work was performed on the quantification of uncertainties of the cost estimates produced with the ACCERT algorithm. Previously, the cost models of the ACCERT algorithm gave deterministic estimates, which can be considered the "expected values" of cost estimates that are, in reality, uncertain. The shape and magnitude of the uncertainties were, however, unknown until the work was performed in 2019. The standard deviation and the functional form of the uncertainty distributions were derived in this work for the first time, for well-executed construction projects, based on actual historical data on input cost uncertainty, such as labor, steel, concrete etc. As an example, it was possible to calculate that the standard deviation of the reference PWR containment's cost is 8.3% of the expected value. Therefore, in order to have a 95% confidence that the actual realized cost of a containment construction will be within the allocated budget, it might be necessary to allocate a contingency of two standard deviations, or of 16.6%. This contingency would be sufficient to cover most of the uncertainty in input costs, which are outside of the control of the constructors.

### 2.3 Development, Deployment, and Implementation Issues

In the Development, Deployment, and Implementation Issues work area, the campaign conducts analyses related to transition from the current U.S. NES to an alternative NES, considering various deployment and implementation scenarios. This includes implementation and application of the TSRA process on NES technologies currently being developed in the NFCSC program, the analyses of nuclear energy in the context of the overall energy system/market, and the enhancement of tools and capabilities for analyses of NES.

#### 2.3.1 Reassessing Methods to Close the Nuclear Fuel Cycle

Since 2012, the SA&I Campaign has been performing a multi-stage assessment of alternate fuel cycles impacts, including an evaluation and screening of 4,400 potential fuel cycles to identify those with the potential for achieving substantial improvements compared to the current fuel cycle and studying how to transition to these "most promising" advanced fuel cycles. This work produced numerous reports over this time period, and it concluded this year with a summary level paper containing over three dozen key findings. Major findings include:

Assuming the continued use of nuclear power in the U.S., transition to a closed fuel cycle based on
continuous recycle is feasible to achieve during the time period when the current fleet of reactors need
replacement. The continuous-recycle fuel cycle includes producing fissile material in fast reactors
instead of obtaining it from mining uranium.

- The transition requires development of a significant inventory of fissile material to initially fuel the reactors. Once recycle is established, the ongoing resource requirements would be a less than 1% of current requirements and high-level waste mass would be only ~5% of current generation for the same level of energy production.
- Two methods for producing the fissile material inventory are from recycling the used fuel from current thermal reactors or the use of High-Assay Low-Enriched Uranium (HALEU). The recycling of thermal reactor used fuel is the traditional approach and used fuel separations facilities are currently operating in France and Russia. However, this study found that the use of HALEU may be more favorable because it is less constrained and may be more economical. The benefits of start-up on HALEU include:
  - o Fewer technologies required to initiate transition
  - Major investments in fuel separation facilities can be delayed
  - o Fuel fabrication costs during initial transition will be lower
  - The system would be more robust and better able to handle disruptions in the recycle system without impacting reactor fuel availability.

The remaining stage of this work will focus on identifying the key technologies required to enable fuel cycle transition. Additionally, researchers will identify the current maturity levels of these technologies, and the R&D activities needed to bring the technologies to the level necessary for constructing demonstration facilities. If desired, R&D will incorporate the TSRA process discussed in the next section.

# 2.3.2 Technology Maturation Planning and Technology Development Roadmap Example Using the Technology and System Readiness Assessment Process

The TSRA process development was completed in 2019 and used to identify and prioritize R&D activities to develop a *Technology Development Roadmap* for a continuous-recycle system using a sodium-cooled fast-spectrum reactors, metallic U/TRU fuel, and electrochemical reprocessing. The 2019 effort included development of a process and reporting format for creating technology maturation plans and rolling those plans up into technology development roadmaps. Major subsystem selection was developed through consultation with the NTDs of the R&D programs associated with reactor, fuels, and reprocessing development. The NTDs selected these subsystems and associated performance objectives primarily to test the TSRA process. While they are compatible, the selected subsystems and performance objectives are not to be construed as an endorsement for any specific fuel cycle configuration. In general, system performance objectives were assumed based on what would be reasonable for a full-scale commercial deployment of the technologies to demonstrate the system by 2040, but would also require use of lower cost advanced materials/technologies that are currently not mature.

A webinar on this work was held in early November 2019 to introduce the TSRA process, provide information on application of the process on the example nuclear energy system, and discuss how the TSRA process could assist with the DOE-NE program integration. About 30 personnel from various DOE-NE offices and national labs participated in the webinar.

# 2.3.3 Load Leveling Through Battery Electric Vehicles and Implications for Electric Power Generation

The emergence of battery electric vehicles (BEV) for transport services, advances in the development of wind and solar photovoltaic energy technologies, and global climate change concerns are converging in a way as to challenge and disrupt the energy system in the U.S. and world. Changes to the daily electricity load curve from the penetration of renewable energy and future projected increases in the daily load from BEV charging could have a large impact on the choice of power technologies and the management of daily electricity demands. A novel approach combining long-term energy simulations from a global integrated-assessment model with an offline analysis of U.S. hourly electricity demand is implemented within the GCAM tool to explore the potential of BEV load flexibility for accommodating greater variable energy penetration while leveling the daily load curve for dispatchable and baseload generation. Results show that the successful deployment of BEVs leading to the electrification of all road transport and flexibility of BEV daily charging has the potential to stabilize and level the daily load curve for dispatchable generation, while enabling greater renewable energy use in the U.S. Flexible operation of dispatchable technologies was minimized and baseload generation provided a greater share of daily power needs.

In a climate change scenario that limits global mean temperature change to 2°C, the daily baseload power output grew to 783 GW by 2100 with flexibility of BEV load demand and 30% renewable energy share of total electricity generation. Flexibility in the daily transport load increased baseload power levels by 20% and reduced the variability in the daily dispatchable power level by 332 GW, as compared to a scenario without BEV load flexibility. These results show that baseload technologies, such as nuclear power, could have a greater role in a future with transport electrification and effective implementation of demand-side management approaches for BEV charging.

#### 2.3.4 OR-SAGE Siting Analyses for Advanced Nuclear Energy Systems

The OR-SAGE dynamic visualization database tool has been further developed in 2019 under the SA&I Campaign to enable additional functionality, including application to fuel cycle facilities, advanced fuel cycle deployment scenarios, and transportation considerations. The expansion tool, Oak Ridge Advanced Fuel Cycle Logistics Environment (ORACLE), will ultimately allow stakeholders to perform interactive parameter and sensitivity studies for the evaluation of fuel cycle facility deployment and optimization. A variety of static results were produced using a specific set of input parameters in order to provide an early demonstration of the potential of the tool. These parameters are based on industry norms and expert judgment. In the future, additional experience and stakeholder input are required to provide the optimum baseline set of criteria. The strength of the ORACLE tool is that numerous alternative scenarios can be quickly generated with associated visualization to provide additional insight into the advanced reactor fuel cycle, its deployment, logistics, and transport. The screening process divides the contiguous United States into 100 x 100 m (1-hectare) squares (cells), applying successive site selection and evaluation criteria (SSEC) to each cell. There are just under 700 million cells representing the contiguous U.S. If a cell meets the requirements of each criterion, it is deemed a candidate area for siting a specific power generation form relative to a reference plant for that power type. Some SSEC parameters preclude siting a fuel cycle facility because of an environmental, regulatory, or land-use constraint. Other SSEC assist in identifying less favorable areas, such as proximity to hazardous operations.

#### 3. ADVANCED FUELS CAMPAIGN

Steven L. Hayes, INL, NTD

#### 3.1 Overview

#### 3.1.1 Mission

The mission of the Advanced Fuels Campaign (AFC) is to perform RD&D activities in support of identifying and developing innovative fuels, cladding materials, and associated technologies to improve the performance and safety of current and future reactors; to increase the efficient utilization of nuclear energy resources; to contribute to enhancing proliferation resistance of the nuclear fuel cycle; and to address challenges related to waste management and ultimate disposal issues.

#### 3.1.2 Objectives

The AFC is responsible for developing and demonstrating innovative nuclear fuel technologies for the Office of Advanced Fuels Technologies. The current priorities for the campaign include:

- 1. Support the industry-led development of Accident Tolerant Fuel (ATF) technologies with improved reliability and performance under normal operations and enhanced tolerance during hypothetical accident scenarios, with implementation of reload quantities of one or more ATF concepts in commercial reactor(s) as early as 2023.
- 2. Lead research and development on innovative fuel and cladding technologies for applications to future advanced reactors, especially fast-spectrum reactors, including reactors for both once-through and recycle applications.
- 3. Develop and demonstrate innovative approaches to accelerate the qualification of new fuels, to include:
  - Close coupling between experiments and advanced fuel modeling and simulation
  - In situ instrumentation of irradiation experiments
  - Microstructure-based mechanistic fuel modeling, informed/validated using advanced characterization of irradiated fuels and materials
  - Accelerated fuel testing
- 4. Collaborate with the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program under NE-5 on the development and validation of multiscale, multiphysics, and increasingly predictive fuel performance models and codes, with particular emphasis on ATF/high burnup fuels for LWRs and metallic fuels for advanced reactors.

#### 3.1.3 Challenges

**LWR Fuels with Enhanced Accident Tolerance.** Improvements will be measured by increased margin to fuel failure, increased coping time to allow for response during an accident to prevent severe damage to the core, and reduced hydrogen generation when the core is uncovered, and the fuels and cladding are in contact with steam.

**Near Zero-loss Fuel Fabrication Processes.** The challenge for fabrication is to lower irretrievable losses of actinide materials to near 0%, whereas current levels are typically on the order of 1-2%. This requires the development of new and more efficient fabrication processes without imposing an economic penalty on fuel fabrication. The objective is to minimize waste generated during fabrication (and potential refabrication) processes and maximize utilization of actinide resources.

Major Increase in Fuel Burnup and Performance over Current Technologies. An increase in fuel burnup is desired for all fuel cycle options. However, the quantitative goals for burnup depend on the reactor type(s) and, more importantly, the selected fuel cycle option(s). In some cases, there are practical and economic limitations to burnup beyond fuel cycle efficiency and technology limitations. Burnup in once-through cycles is limited by the initial enrichment constraints and cladding material properties. Burnup for fuels used in a full recycle scenario may be limited by reactor physics, storage, and/or disposal constraints after the discharge of spent fuel. Another important consideration in increasing fuel burnup is to ensure near zero-failure of fuels in-reactor, a standard that industry is striving to achieve at current burnup limits. Quantitative bounds for burnup objectives under various fuel cycle scenarios will be developed as the program progresses and fuel cycle scenarios are defined.

#### 3.1.4 Major R&D Activities

Advanced LWR Fuels with Enhanced Accident Tolerance. Since 2012, development and demonstration of LWR fuels with enhanced accident tolerance (i.e., Accident Tolerant Fuels) has been the highest priority, near-term activity of AFC. The three DOE-sponsored industry teams (led by Westinghouse, General Electric, and Framatome) are responsible for leading the development and qualification of their independent ATF concepts. The activities of the three industry teams are directly managed by DOE-NE, which means they are not accountable to AFC technical leadership at the laboratories. The role of the laboratories is to: (1) develop and maintain needed fuel testing/qualification infrastructure, (2) perform independent testing and evaluation of all ATF concepts, and (3) support individual industry funding opportunity announcement teams at their request, and supported by funding under their direction.

Advanced Reactor Fuels. Development and testing of fuels and materials for application in future, advanced reactors (especially fast-spectrum reactors) is the second, longer-term activity of AFC. Primary R&D activities include development and testing of fabrication technologies to improve yield and minimize waste generation; characterization of fuels and documentation of their properties in archival handbooks; fuels and materials testing in the Advanced Test Reactor (ATR), High-Flux Isotope Reactor (HFIR), and Transient Reactor Test (TREAT) with a view to demonstrating reliable performance at high temperature and/or to high burnup; and development/demonstration of combined testing and modeling methodologies that could accelerate the development and qualification of new fuels.

Capability Development. Major activities in the area of capability development included characterization instrument development and installation in hot cells at the Irradiated Materials Characterization Laboratory, design and fabrication of generic and reusable test trains for transient experiments in the TREAT Facility, conceptual design of I-Loop and supporting systems for boiling water and ramp testing of advanced LWR fuels in ATR, implementation methods for advanced instrumentation/sensors to be used with fuel experiments in ATR and the TREAT Facility, and mechanistic model development and implementation into BISON (in collaboration with NEAMS).

International Coordination and Collaboration. Researchers from AFC participated in historic international collaborations with Korea, France, Japan, the European Atomic Energy Community (EURATOM), and the Organization for Economic Cooperation and Development-Nuclear Energy Agency (OECD-NEA), with new collaborations with the United Kingdom and Canada started in 2018. These collaborations are managed through a combination of participation in Generation IV Global International Forum projects, International Nuclear Energy Research Initiative (INERI) projects, and participation in bilateral and trilateral government-to-government agreements. During 2019, active work was executed in collaboration with KAERI (Korea), CEA (France), JAEA (Japan), EURATOM (INERI), and OECD-NEA, with increasingly mature interactions with the UK and Canada on potential areas of future collaboration.

#### 3.1.5 Major Achievements

A comprehensive summary of campaign accomplishments during 2019 can be found in the *Advanced Fuels Campaign 2019 Accomplishments Report* (which is available for download at: https://nuclearfuel.inl.gov/afp/SitePages/Home.aspx). Of particular note are the following major achievements and their significance:

- The Halden Gap Assessment Final Report with recommendations to DOE-NE on strategies and investments needed to support testing and qualification of industry-led ATF concepts without the Halden Reactor was issued in December 2018.
- Uranium silicide (U<sub>3</sub>Si<sub>2</sub>) fuel pellets fabricated at INL (Figure 4) were incorporated into fuel rods as part of a Westinghouse lead test assembly (LTA) loaded into the Byron commercial power reactor, which has been operating at full power since April 2019; this represents the first DOE-sponsored commercial LTA that included a new fuel form.
- Fundamental understanding of uranium silicide fuel was extended through temperature-dependent neutron diffraction measurements, thermo-mechanical property measurements, thermodynamic evaluations of U<sub>3</sub>Si<sub>2</sub> hydride reactions, and new fabrication and characterization studies on doped UO<sub>2</sub> fuel concepts were initiated.
- Fabrication trials on reduced-diameter fuel pellets (UO<sub>2</sub> and U<sub>3</sub>Si<sub>2</sub>) were successful, setting the stage for possible accelerated testing in ATR using the Fission Accelerated Steady-state Test (FAST) approach.
- Fracture toughness measurements using small-scale cantilever beam testing were performed using unirradiated UO<sub>2</sub>. They were found to be in good agreement with data obtained using conventional testing. This methodology will be extended to irradiated fuel specimens in the future.
- Variations in minor alloying constituent levels were explored in new heats of FeCrAl cladding resulting in improved properties. Improved measurements of hydrogen diffusion in FeCrAl cladding

- were obtained. Tube production and characterization of oxide-dispersion strengthened FeCrAl variants were initiated, and the FeCrAl Properties Handbook was updated and issued.
- Prototypic testing of Westinghouse and Framatome ATF concepts in ATR's 2A Loop (ATF-2) continued throughout 2019, and test fuel rodlets achieved burnups ~10 GWd/MTU by year end. The ATF-2 Loop continues to be the only in-reactor option in the western world for testing of ATF concepts under prototypic PWR conditions.
- A major ATF-2 test train design modification for BWR-sized rodlets was completed, and 12 new test
  rodlets from General Electric (GE) are to be introduced into ATF-2 during the outage preceding ATR
  Cycle 166B (Figure 5).
- Design of a new 9-pin tier for ATF-2 was completed, which could effectively increase the rodlet capacity of the ATF-2 test train by 50% (a recommendation included in the Halden Gap Assessment).
- Uranium silicide fuels in ATF-1 (Figure 6) reached burnups of ~40 GWd/MTU, the equivalent of two full cycles in a commercial power reactor; interim examinations to date have indicated that these silicide fuels exhibit very low swelling and fission gas release, which is a significant improvement over conventional fuels under normal operations, in addition to benefits they promise to provide under accident scenarios.
- Conceptual design of ATR I-Loops as recommended in the Halden Gap Assessment was initiated, with the objective of providing prototypic boiling water reactor (BWR) test conditions, ramp testing, and run-to-failure testing capabilities needed to qualify ATF concepts without the Halden Reactor.
- A series of commissioning tests were completed in the TREAT Facility using the separate effects test holder (SETH) irradiation capsule (Figure 7), demonstrating the function of all critical systems. These were the first transient fuel tests conducted in the U.S. in ~30 years and represent a significant leap forward in experimental technology.
- The development and deployment of the MARCH-SERTTA submodule was used to conduct the first fueled reactivity-initiated accident (RIA) experiments in the TREAT Facility in a water environment, and the first RIA experiments were performed on an unirradiated ATF concept (U<sub>3</sub>Si<sub>2</sub>).
- Conceptual design of the new FAST irradiation experiment was completed, which will use miniaturized fuel rods to accelerate burnup accumulation by as much as a factor of 10, greatly reducing the time required for reactor testing.
- FAST fabrication trials on miniature fuel rodlets were successfully completed this year (
- Figure 8), including rodlets with integral Zr, V liners; the first FAST experiments with metallic fuel rodlets are scheduled for insertion in ATR during 2020.
- The first MiniFuel irradiation in HFIR of both coated and uncoated nitride microspheres was completed and post-irradiation examination was initiated. This capsule-based, separate effects approach to fuel testing promises to provide fundamental data that will be valuable to advanced fuel modeling efforts. Uranium silicide (U<sub>3</sub>Si<sub>2</sub>) specimens were fabricated at Los Alamos National Laboratory (LANL) and shipped to Oak Ridge National Laboratory (ORNL) for a subsequent MiniFuel irradiation in HFIR to begin in 2020.
- Machining capabilities for transuranic-bearing metallic fuels were developed and deployed in the Fuel Manufacturing Facility in order to demonstrate many of the innovative design features developed in

- recent years, including sodium-free U-Pu-Zr metallic fuels as an advanced driver fuel for the VTR. The first two annular metallic fuel slugs of U-20Pu-10Zr were produced (Figure 6).
- Extensive characterization studies were conducted on metallic fuels with additives (Pd, Sn) for lanthanide fission product immobilization, which are showing promise toward increasing the allowable cladding temperatures in metallic fuel pins. A major update to the Metallic Fuels Handbook was issued, incorporating a considerable amount of new data generated by the campaign.
- Good progress was made on the processing and characterization of nano-structured ferritic alloys, with
  a view to developing a new cladding for sodium fast reactors capable of higher temperature operation,
  and experimental fabrication of oxide-dispersion strengthened steels by extrusion/pilgering continued.
- The first thermal property measurements on U-10Zr metallic fuels irradiated to over 10% burnup in the Fast Flux Test Facility were obtained in the new Thermophysical Properties Cell at the Irradiated Materials Characterization Laboratory at INL.
- The Integrated Recycling Test (IRT) continued irradiation in cadmium-shrouded positions in the ATR throughout 2019. The test contains metallic fuels fabricated remotely using actinide materials recovered from spent fuel. Data from this test will provide important information on the performance of recycled metallic fuels. Peak fuel burnup was nearing 3% at year end.
- Considerable enhancements were made to the BISON fuel performance code for simulations of metallic fuels, including the addition of a new mechanistic fuel swelling model, which was used to investigate the deformation behavior of Na-free, slotted metallic fuel concepts (Figure 10).



Figure 4. Uranium silicide fuel pellets fabricated at INL and included in Westinghouse's Byron LTA.

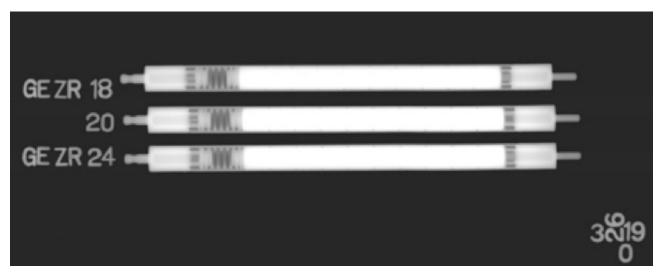


Figure 5. Radiograph inspection of GE rodlets supplied for introduction into the ATF-2 test train.



Figure 6. ATF-1 basket loaded with industry-supplied test rodlets to be irradiated in the ATR.



Figure 7. Post-transient neutron radiography of SETH capsules showing fuel failure threshold for traditional UO<sub>2</sub>/Zircaloy fuel rods.



Figure 8. Finished FAST (surrogate) rodlets in one-half diameter geometries.



Figure 9. The first U-Pu-Zr metallic fuel slugs fabricated in annular (a) and slotted (b) geometries.

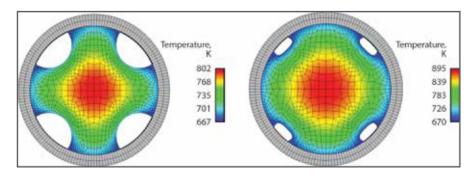


Figure 10. BISON simulations of the swelling behavior of slotted metallic fuel.

#### 3.1.6 Major Activities Planned for 2020

- Continue coordination and integration of activities of ATF industry teams (Westinghouse, General Electric, and Framatome) with that of the laboratories.
- Fabricate UO<sub>2</sub> pellets with both standard and large grain sizes to support accelerated burnup tests being planned in ATR.
- Perform ion irradiations of FeCrAl alloy C26M up to 15 dpa.
- Fabricate new ATF-2 Tier 5/6 and consolidate existing and new Westinghouse-supplied ATF rodlets under irradiation in ATR.
- Complete the design of a Pellet-Cladding Mechanical Interaction (PCMI) test for SiC/SiC cladding to be irradiated in HFIR.
- Complete the post-irradiation examination and assessment of UN kernels irradiated as part of the first MiniFuel capsule experiment in HFIR.
- Perform the MARCH-SERTTA commissioning tests in the TREAT Facility (~10 transients), perform transient tests on ATF concepts using the SETH capsule in the TREAT Facility (~transients), and complete the final design of the Super-SERTTA test train.
- Complete fabrication of metallic fuel rodlets for the FAST-1 experiment and begin irradiation in ATR.
- Prepare MiniFuel test of U-Zr metallic fuel specimens for irradiation in HFIR.
- Issue summary report on mechanical testing of fast reactor cladding materials irradiated to >30 dpa.
- Complete preparations for transient testing in the TREAT Facility using the Temperature Heat-sink Overpower Response (THOR) device, and complete conceptual design of first sodium loop experiment for the TREAT Facility.
- Continue cooperation with NEAMS to improve, enhance, and validate BISON for simulations of ATF/high burnup fuels for LWRs and metallic fuels for advanced reactors.
- Procure a re-fabrication station for irradiated fuel rods to be installed at INL for the support of instrumented experiments in ATR and/or the TREAT Facility.
- Complete design and fabrication of a new ATR Top Head Closure Plate with additional feedthroughs to support future I-Loops.

#### 4. JOINT FUEL CYCLE STUDY

Ken Marsden, INL, NTD

#### 4.1 Overview

The Joint Fuel Cycle Study (JFCS) program is exploring the technical and economic feasibility and nonproliferation acceptability of spent fuel management options with international program stakeholders. Electrochemical recycling is one technology being evaluated, and this process utilizes dry processes that allow collection of recovered uranium and group collection of a uranium-transuranium product. Although often conceived for recycling of irradiated metallic fuels, head-end processes can be applied to allow recycling of actinoid elements from irradiated oxide fuels.

The JFCS is a schedule-driven activity of 10-year duration beginning in 2011. The study is divided into three phases, each of which have an area of primary emphasis. The first phase was 2 years in duration and focused on the Laboratory-Scale Feasibility Study to verify the scientific feasibility of Electrochemical recycling at small scale. The second phase was 5 years in duration (2013–2017) and primarily emphasized demonstration of integrated process operations and recovery of sufficient fuel material to enable recycled fuel fabrication. The third phase is 3 years in duration (2018–2020) and focuses on evaluation of recycled fuel fabrication and its irradiation performance, processing of irradiated LWR fuel, and demonstration of waste form processes.

Key outcomes for the program in 2019 include:

- Completion of two process tests with approximately 2 kilograms of irradiated LWR fuel from the BR3 and North Anna reactors
- Cooldown, evaluation, and restart of the molten salt system used for oxide reduction which had operated at 650°C continuously for approximately 32 months
- Completion of a series of waste form studies using irradiated materials from JFCS testing
- Loading, transportation, and receipt of approximately 35 kilograms of irradiated LWR fuel to be used as testing feedstock, discharged primarily from the Dresden plant
- Initiation of testing and process demonstrations in a five-batch campaign with the irradiated Dresden LWR fuel at a 4-kilogram batch size.

An image of irradiated LWR fuel received in the transportation cask is provided in Figure 11, prior to transfer into the Hot Fuel Examination Facility. Figure 12 is a photograph of an irradiated stainless-steel-based metal waste form produced to sequester noble fission products. Figure 13 is an image of the irradiated Dresden LWR fuel following removal from the zircaloy cladding.

Due to program sensitivity, additional information is available upon request.



Figure 11. Irradiated LWR fuel in the NAC-LWR cask upon receipt at the Hot Fuel Examination Facility (HFEF).

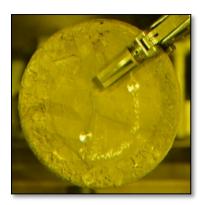


Figure 12. Stainless-steel-based metallic waste form produced using irradiated fission products from JFCS testing.



Figure 13. Irradiated LWR fuel from the Dresden reactor following removal from zircaloy cladding.

# 5. MATERIAL RECOVERY AND WASTE FORM DEVELOPMENT CAMPAIGN

Terry Todd, INL, NTD John Vienna, Pacific Northwest Nuclear Laboratory, Deputy NTD

The MRWFD Campaign develops advanced separation and waste processing technologies to support the various fuel cycle options. Although research is performed to support a range of potential fuel cycles, the focus is on the most promising fuel cycles evaluated in the *Nuclear Fuel Cycle Evaluation and Screening – Final Report*, October 2014, actinide recycling. The campaign also performs activities to enable advanced nuclear reactors by providing High-Assay Low-Enriched Uranium (HALEU). This section provides a highlight of the results of R&D efforts performed within the MRWFD Campaign in 2019. Each subsection contains a high-level overview of the activities and key results, produced during the year.

This section briefly outlines the campaign mission, objectives, and challenges, and highlights key technical accomplishments made during 2019. The campaign continued to utilize an engineering-driven science-based approach to maintaining relevance and focus.

The MRWFD Campaign management and integration activities included international collaboration activities (primarily focused on bilateral and multilateral collaborations with France, Japan, the European Union, and the International Atomic Energy Agency [IAEA]), integration of MRWFD Campaign activities with other NFCSC campaigns, (primarily Advanced Fuels, Used Fuel Disposition and Fuel Cycle Options, and Material Protection, Control, and Accountability Technologies).

Technical accomplishments are reported under the following R&D categories:

- Process chemistry and integration
- Integrated CoDCon demonstration
- Sigma team for off-gas capture and immobilization
- Advanced Wasteform development and performance
- ZIRCEX/HALEU recovery
- Domestic electrochemical separation technologies



#### Mission

Develop advanced fuel cycle separation and waste management technologies that improve current fuel cycle performance and enable a sustainable fuel cycle, with reduced processing, waste generation, and potential for material diversion.

#### 5.1 Mission

The MRWFD research team applies expertise and technical capabilities to a wide array of applications. This campaign now also leverages its expertise by working with others in areas such as environmental remediation, national science, security missions, as well as civilian nuclear applications. The mission of the MRWFD Campaign is to:

Develop advanced fuel cycle separation and waste management technologies that improve current fuel cycle performance and enable a sustainable fuel cycle, with minimal processing, waste generation, and potential for material diversion.

### 5.2 Objectives

- 1. Develop and demonstrate options for *enabling* advanced reactors by making HALEU feed material available.
- 2. Develop and demonstrate material recovery technologies targeting high-value used nuclear fuel (UNF) that would be generated from advanced reactors (e.g. with enrichments ranging from 10–19%).
- 3. Develop and demonstrate crosscutting technologies that support any advanced fuel cycle including:
  - Capture and immobilization of volatile fission products (e.g. I, Kr, tritium, etc.) and waste treatment options optimized around performance and cost.
- 4. Maintain technical expertise and research capabilities in the DOE complex for nuclear fuel cycle RD&D.

### 5.3 Challenges

- Separate minor actinides from lanthanides in both aqueous and molten salt media.
- Capture and immobilize off-gas constituents of used fuel, including iodine, krypton, tritium, and potentially carbon, in a cost-effective manner.
- Develop separation technologies and waste forms relative to the types of fuels being processed, the types of fuels being fabricated, and the reactors used to burn recycled fuels.
- Achievement of advanced separation and immobilization processes in a cost-effective manner.
- Predict performance of waste forms with lifetimes measured in units of millions of years.

### **5.4 2019 Progress**

#### 5.4.1 Process Chemistry

This activity supports evaluation of solvent degradation mechanisms and a fundamental understanding of radiation chemistry in nuclear fuel cycles, specifically in used fuel processing operations. Understanding the mechanisms and rates for solvent degradation under relevant process conditions is essential for any process exposed to high radiation fields. The fundamental understanding of free radical formation and their effects on the process, including the reaction kinetics of the free radicals, enables development of predictive models of radiation damage. This activity also supports development of novel metal chelators to improve methods of differentiating trivalent actinides from trivalent lanthanides, including developing a fundamental understanding of the thermodynamic structure-function relationships for radiometal chelation (as shown in Figure 14). This task is supported by organic synthesis and fundamental modeling efforts. The MRWFD Campaign has an active collaboration in the European Union Generation IV Integrated Oxide Fuels Recycling Strategies (GENIORS) program through an INERI project.

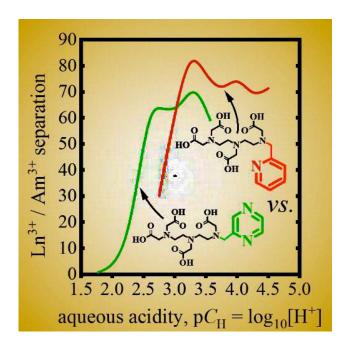


Figure 14. Showing the change in separation factor between Ln<sup>3+</sup> and Am<sup>3+</sup> as a function of pH, where the lower pH values for the green ligand enable the separation to be performed under more robust and stable conditions.

#### 5.4.2 CoDCon Flowsheet Demonstration

The CoDCon flowsheet demonstration was established to quantify, using laboratory-scale equipment, the accuracy and precision to which a specific uranium-to-plutonium (U/Pu) ratio can be achieved in the Pucontaining product from a tributyl phosphate-based nuclear fuel recycling flowsheet. For the purpose of this project, the target U/Pu-mass ratio is 7/3 (2.33), in both the mixed U/Pu nitrate stream from the solvent extraction and in the final mixed oxide fuel product. The uncertainty associated with achieving a specific target U/Pu ratio will be established. Another major objective of the project is to demonstrate optical spectroscopic techniques for real-time monitoring of the concentrations of key components (e.g., Pu, U, and HNO3) in the process solutions. The monitoring capability is viewed as critical to achieving the first stated objective of controlling the process to achieve a product with a U/Pu-mass ratio of 7/3.

In 2018, three CoDCon flowsheet tests were performed in a shielded glovebox using a simplified simulant feed (U and Pu in nitric acid). Results from the three tests were very promising. It appears that using flow control of the tributyl phosphate stream into the stripping section, the U/Pu ratio can be controlled to within one per cent variation (see Figure 15). A fourth test was performed in 2019 using a more complete simulant (one containing most of the chemical constituents of UNF). Although the average composition of the U/Pu product in the fourth test was within 0.3% of the target ratio, control of the product ratio control was not as good as with the simplified simulant. The standard deviation in the U/Pu ratio over the course of the experiment was 2.5%. Environmental conditions might explain the drifting during the fourth test, since there were substantial changes in the ambient temperature throughout the course of the test. Adjustments have been made to the flowsheet and monitoring chemometric software, and a fifth test with a full simulant is planned for 2020.

#### Relative U/Pu in the Run 3 Product

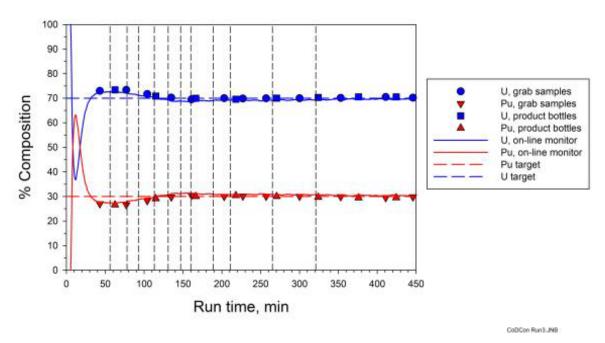


Figure 15. Ratio of U and Pu in CoDCon test 3 product stream.

#### 5.4.3 Sigma Team for Off-Gas Capture and Immobilization

This activity is needed to enable any new fuel treatment facility to meet current regulations. The capture of iodine at very high decontamination factors is required and iodine has a very long half-life, so immobilization is important to reducing the source term in a geologic repository. Krypton (Kr) capture will be needed if processing fuel less than roughly 30 years old. Tritium may also require capture if removed from fuel at the head-end. It is very important to understand the behavior of the entire off-gas system to avoid cross-contamination of sorbents (e.g., iodine on tritium or krypton sorbents). Two significant accomplishments were completed in 2019: (1) an integrated gas treatment system was designed, built, and tested to remove iodine and tritium from an advanced tritium-pretreatment operation and (2) a new engineered form of metal organic frameworks for room temperature krypton capture was invented and tested.

#### 5.4.4 Advanced Waste Forms Development and Performance

These activities are necessary for the immobilization of waste streams from the advanced fuel cycle options, including advanced aqueous and electrochemical processes. These waste forms are designed for significant reduction in implementation cost and risks and/or significant improvements in long-term performance in disposal. Lower costs and risks can be achieved by simpler processes, higher waste loadings, and removal of streams through purification and/or recycling. Higher performance can be achieved by more durable waste form phases for high-level waste and radioiodine. Results of some waste form activities are shown in Figures 16 through 18 for electrochemical salt waste, iodine waste forms, and ceramic waste forms, respectively.

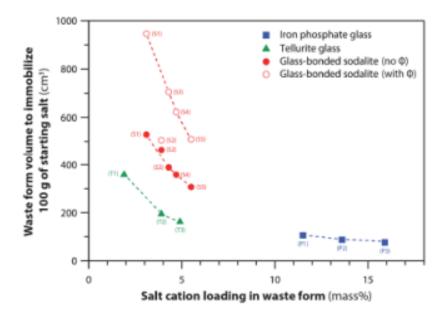


Figure 16. Comparison of final waste form volumes between different proposed electrochemical salt waste forms showing more than an order of magnitude reduction for iron phosphate glass compared to glass bonded sodalite (from Riley 2019).

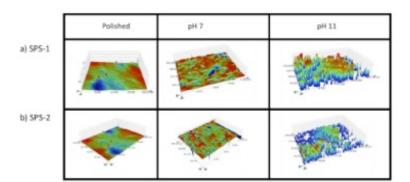


Figure 17. Optical profilometry of example iodine waste forms under different pH conditions (Asmussen et al. 2019).

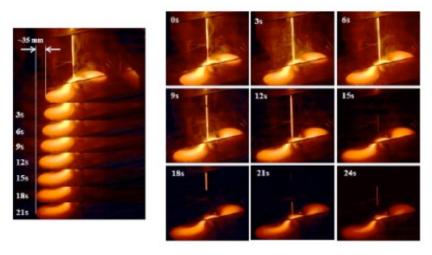


Figure 18. Time-lapsed photograph of glass ceramics melter test pour (Amoroso 2018).

#### 5.4.5 ZIRCEX Demonstration

The ZIRCEX Material Recovery Pilot Plant (MRPP) construction and operability testing were completed in 2019. A number of issues (heating system, valves, etc.) had to be redesigned and/or replaced. Functional acceptance testing was completed, and the system was deemed operable after a management self-assessment was performed. The MRPP will begin testing zirconium samples in early 2020, followed by testing of aluminum samples, and, finally, unirradiated naval fuel.

#### 5.4.6 Hybrid ZIRCEX Engineering-Scale Demonstration

In the Hybrid ZIRCEX process, conventional UNF receipt and handling techniques were used to feed Step 1, where the fuel's zirconium or aluminum cladding is reacted and volatilized with chlorine-based gas in a

fluidized bed at elevated temperature. The highly-enriched uranium (HEU) remains in the fluidized bed until it is elutriated and collected in another vessel. The recovered HEU is then dissolved in nitric acid and fed to the solvent extraction process in Step 2. Solvent extraction separates and purifies the uranyl nitrate. The liquid product (HEU) stream is blended as a liquid in Step 3 with low-enriched uranium (LEU), natural uranium (NU), or depleted uranium (DU) to form HALEU. The liquid HALEU is then denitrated and solidified to form an oxide (or metals, carbides, etc., with other proven technologies) suitable for fuel fabrication. The fission products are vitrified at volumes 300 times less than achieved with traditional dissolution methods because the cladding is removed in Step 1 and disposed of as lowlevel waste (Figure 19).

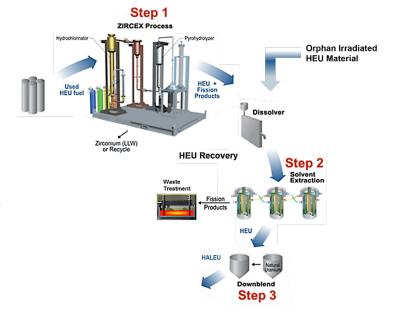


Figure 19. Hybrid ZIRCEX Process.

Demonstration of the INL Hybrid ZIRCEX Process will require progressively larger demonstrations, culminating in an engineering-scale demonstration of all three steps, which recover HEU from UNF. Over 20 engineering studies were completed by INL Engineering to define the scale demonstration, resulting in issue of functional and operational requirements (F&ORs) to support the start of conceptual design.

The first of these studies was an analysis of alternative site locations in which two acceptable locations were identified using engineering techniques, consistent with best recommended practices. The east side of the Materials and Fuels Complex was recommended based on final scores of the acceptable locations as shown (Figure 20).

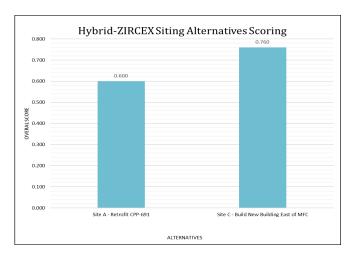


Figure 20. Hybrid ZIRCEX Alternative Scoring systems.

Another notable study, ECAR-4705 "Hybrid ZIRCEX Reprocessed Fuel Product Composition," predicted the composition of the HALEU product from the Hybrid ZIRCEX process using the Fluorinel Design Basis Fuel Element and separations factors from varied experimental reports and Argonne Model for Universal Solvent Extraction (AMUSE) modeling. The composition was compared to three fuel specifications. Fuel specifications were met, apart from radioactive activity and the concentrations of samarium and tin, which are likely the consequence of uncertainty in the separation factors. Planned demonstrations in subsequent years will reduce the uncertainty in the separations factor.

Additionally, a National Environmental Policy Act of 1969 strategy was developed to build and then conduct the engineering-scale demonstration with an environmental assessment to compliantly address the policy requirements. After successful demonstration of uranium recovery and conversion from zirconium-and aluminum-clad UNF, an environmental impact statement will be required if DOE decides to use the demonstration after the demonstration period.

Based on the engineering studies, it was determined that the engineering-scale demonstration could be built and operated with Operating Funds. A world-class Architectural-Engineering firm was contracted for conceptual design of the engineering-scale Hybrid ZIRCEX Demonstration starting in Fall 2019, confirming that the engineering-scale Hybrid ZIRCEX Demonstration is progressing on pace to demonstrate the recovery of HEU from UNF by 2025.

#### 5.4.7 Hybrid ZIRCEX/HALEU EBR-II Polishing

A laboratory-scale glovebox system was developed and installed for the polishing of contaminated HALEU materials, to demonstrate they could be cleaned to meet fuel specifications. The system included a dissolver, 16 stages of Robatel mixer-settlers, product precipitation vessel, and a furnace for calcining the uranium product. The first demonstration will be a 1.7 kg uranium metal ingot of EBR-II product. The metal will be dissolved, purified by solvent extraction, and the product precipitated and calcined to uranium oxide. A view of the system is shown in Figure 21.

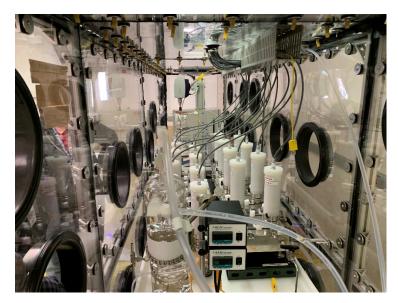


Figure 21. HALEU polishing equipment inside glovebox.

Researchers at INL performed preliminary dissolution experiments of DU metal and a 123-gram disk of EBR-II material in hydrofluoric acid to determine dissolution temperature and acid composition for dissolution of EBR-II HALEU. Preliminary dissolution experiments are in preparation for dissolution of a 1.7 kg EBR-II HALEU specimen in a laboratory-scale HALEU polishing demonstration. A 1.7 kg EBR-II ingot was transferred to CFA-625 from the Fuel Conditioning Facility (FCF) in container DCR-004, as shown in Figure 22. The ingot will be used to determine dissolution parameters and demonstrate the removal of chemical and radioisotope impurities ("polishing") of the EBR-II HALEU product at lab scale.

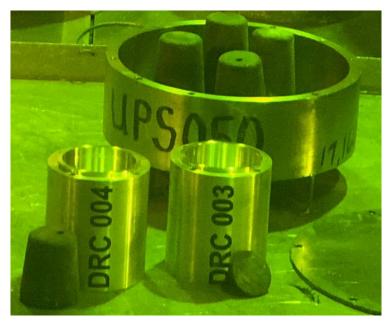


Figure 22. 123 g and 1.7 kg EBR-II product.

#### 5.4.8 Domestic Electrochemical

This activity is developing technologies to potentially enhance performance and reduce waste volumes in the treatment of metallic fast reactor fuels for TRU recycling. In 2019, design and construction of a modified, kg-scale U/TRU code position system using a high-current-density solid cathode was completed. Studies were performed to identify solutions for technical gaps in Electrochemical recycle technology. Additionally, planning for future testing of the solid cathode for U/TRU recovery from actinide-containing salts was continued.

## 5.5 Key 2019 Deliverables

The following 2019 deliverables were completed.

- Completed an analysis of alternative sites for the engineering-scale Hybrid ZIRCEX Demonstration
- Completed support of DOE's Report to Congress describing a plan and cost profile for developing HALEU
- ECAR-4705 "Hybrid ZIRCEX Reprocessed Fuel Product Composition" predicted the composition of the HALEU product from the Hybrid ZIRCEX process
- Completed dissolution testing of 124-gram disk at the Materials and Fuels Complex analytical laboratory to define dissolution parameters for laboratory-scale polishing
- Completed transfer of 1.7 KG (total U) EBR-II product to Central Facilities in preparation for laboratory-scale polishing
- Completed assembly of equipment in glovebox to polish EBR-II metal product to meet fuel fabrication specifications
- Demonstrated the ability to recover both the iodine and tritium that would be released during tritium pretreatment of UNF in a manner that the releases from the fuel can be quantified; system also demonstrated the combined recovery system could operate in any of the three proposed tritium-pretreatment modes (once-through air or O<sub>2</sub>, recirculating air or O<sub>2</sub>, or recirculating NO<sub>x</sub>)
- Completed one CoDCon flowsheet test with full dissolved fuel simulant
- Completed functional acceptance testing and a management self-assessment for the ZIRCEX Material Recovery Pilot Plant at INTEC CPP-653; testing began in early 2020
- A glass composition for the immobilization of electrochemical processing salt high-level waste was
  developed with an order of magnitude improved chemical durability and more than an order of
  magnitude higher volumetric loading compared to the baseline glass bonded sodalite for the same
  waste stream
- Simulated the effects of ballistic radiation damage on waste glass as part of a joint DOE-CEA effort
- Developed suite of tests to compare the long-term durability of iodine waste forms showing significant differences between currently proposed forms
- Performed scaled melter test to demonstrate the continuous fabrication of an advanced glass ceramics waste form
- Completed design and construction of equipment or demonstrated engineering-scale feasibility of solid electrode U/TRU co-deposition.

## 5.6 Summary

The MRWFD Campaign continues to make significant progress toward the development and understanding of nuclear materials recovery, waste form development, waste form performance, and nuclear materials processing. These contributions are recognized worldwide and have resulted in several publications in prestigious journals and invitations to present at international conferences.

# 6. MATERIAL PROTECTION, ACCOUNTING AND CONTROL TECHNOLOGIES PROGRAM

Mike Browne, LANL, National Technical Director

#### 6.1 Overview

The mission of the Materials Protection, Accounting and Control Technologies (MPACT) campaign is to develop innovative technologies and analysis tools to enable next-generation nuclear materials management for existing and future U.S. nuclear fuel cycles.

In 2019, the MPACT campaign continued to focus on the Milestone 2020 activities. Milestone 2020 is a demonstration of Safeguards and Security by Design (SSBD) applied to a virtual Electrochemical (Echem) recycling facility validated through modeling and experimental data obtained in coordination with DOE-NE and DOE-NNSA programs. The goal of this effort is to (1) develop a material control and accounting (MC&A) approach for a domestic Echem facility, and (2) utilize this experience to develop a general framework/methodology for MC&A systems for future nuclear fuel cycle facilities. Applying this approach to DOE-NE fuel cycle activities (e.g. recycling, advanced reactors) will facilitate the identification and resolution of potential MC&A issues early in the fuel cycle development.

To support the Milestone 2020 effort, MPACT researchers refined and updated Echem-focused models based on operational data from the JFCS campaign, including the physical models for the unit processing steps, the safeguards models that track nuclear material through the facility and evaluate safeguards performance, and the security model to assess vulnerabilities with the virtual facility. Researchers also made significant advancements in MC&A technologies for material accountancy and process monitors. In 2019, most of these R&D efforts switched focus from laboratory development to field installations and tests at INL, ANL, and LANL facilities.

Finally, MPACT personnel performed a high-level assessment of MC&A needs for identified FCRD. The goal of this assessment was to take a strategic look at NE fuel cycle developments and identify areas where MPACT may be able to address MC&A needs of these developments based on MPACT technologies, modification of MPACT technologies, or application of the Milestone 2020 approach.

#### 6.1.1 Objectives of MPACT Campaign

The MPACT campaign vision is to support an economically competitive fleet of domestic advanced reactors and fuel cycle facilities meeting current and future MC&A requirements. The campaign strategy is to:

- 1. Develop innovative technologies, analysis tools, and advanced integration methods
- 2. Coordinate and engage with government and industry stakeholders early in the technology development process.

We enact this strategy through technical research and development activities to explore & incorporate coordinated SSBD concepts in the following general areas:

- MC&A technologies development and demonstration
- Safeguards and security systems analysis tools to improve MC&A efficiency and effectiveness

- Advanced integration methods to apply effective combinations of technology and analysis tools
- Technical assessments to support MC&A of advanced fuel cycle concepts and approaches
- Guidelines for SSBD and potential application to new facility concepts.

#### 6.1.2 Drivers for MPACT Campaign

In 2019, the MPACT campaign pursued activities that directly support or could eventually support, MC&A needs for DOE/NE efforts in the following areas:

- Material Recovery and Waste Form Development (MRWFD)
  - Material recycling (advanced aqueous processes)
  - Waste forms
- VTR
  - Potential MC&A technology test bed
- Electrochemical Recycling
  - Input, in-process, and output NMA/safeguards and security
- Molten Salt Reactor (MSR)
  - Solid and liquid fuel NMA/safeguards and security
  - Modeling coordination
- Advanced Reactor Technologies (ART)
  - MC&A technology development with potential application to:
    - Fast Reactor NMA/safeguards and security
    - Recycled Used Fuel (UF)
    - Special purpose applications: small/microreactors.

Specifically, in 2019, the MPACT campaign pursued activities to:

- Develop and conduct field test plans for supporting technologies with emphasis on:
  - Milestone 2020 (Echem)
  - R&D into cross-campaign beneficial MC&A technologies
- Performed sensitivity studies for Echem safeguards and security performance
- Developed physics-based time-dependent signatures to guide advanced monitoring technology development (ongoing)
- Developed an advanced integration approach for disparate data
- Developed and test integrated an approach for Echem safeguards and security
- Developed and test integrated approaches for safeguards and security of advanced reactors and processes

#### 6.1.3 Key 2019 Deliverables

- Milestone 2020: Completed the development of an Echem virtual facility definition with operational parameters and NMA/safeguards and security target values
- Exploratory R&D: Completed a measurement campaign and end-user assessment for the MPACT-relevant microcalorimetry technology
- MC&A Advanced Fuel Cycle Scoping Completed an evaluation and assessment of NMA/safeguards and security for identified FCRD.

## **6.2** Major Research and Development Activities

#### 6.2.1 Milestone 2020 Safeguards and Security by Design – Echem

SSBD is a methodology and discipline for integrating next-generation MPACT considerations into the design of nuclear facilities from the very earliest stages. The goal is to identify innovative process and facility design features that maximize the effectiveness and efficiency of safeguards and security, and to work with the design team throughout the design process to introduce such features as appropriate, thereby minimizing the need for costly retrofits. The MPACT team selected Electrochemical recycling (Echem) as the test case for application to advanced FCRD in coordination with the Material Recovery and Waste Form campaign and JFCS. Advanced concepts and approaches, analysis tools (Echem safeguards models, Echem security models), and instrumentation (oxide reduction [OR] and electrorefiner [ER] voltammetry probes, actinide sensor, bubbler, U/TRU Pu measurement system) are being developed and applied in an integrated manner to optimize the overall system effectiveness. This ambitious approach starts with first principle models to calculate the flow of material in the Echem facility, a safeguards and security model to evaluate nuclear material accounting and control (NMAC) effectiveness, and integrated modeling analysis of measurement systems to provide realistic input to the safeguards and security model (Figure 23). The models are validated using available experimental data, and data from MPACT-funded technologies are used as measurement inputs.

## Virtual Facility Distributed Test Bed **HIGH FIDELITY CAPABILITIES** SYSTEMS LEVELS MODELS **KEY METRICS 3D Security Model** Consequence Models **Facility Layout** Radiation Signatures Safeguards Model (SSPM) Measurement SEID (σ,....) **Technologies Probability of Detection Measurement Models Experimental Data** Statistical Methods Flowsheet Model (AMPYRE) Unit Operation Models

Figure 23. Advanced integration modeling approach for Echem.

#### Modeling and Simulation for Echem Safeguards and Security:

Milestone 2020 Virtual Test Bed: One of the motivations for the Virtual Test Bed is recognition that safeguards demonstration facilities in the U.S. do not exist and would be costly to implement. However, a number of experimental test beds are available at various laboratories and universities that can provide safeguards, security, or measurement data for specific applications. These data are utilized by systems-level modeling capabilities to develop next-generation safeguards and security designs. In this manner, SSBD approaches can be tested in distributed experimental capabilities which are tied together through virtual computer models.

The Virtual Facility Distributed Test Bed concept is outlined in Figure 23. The center column of the figure highlights three systems-level modeling capabilities. These virtual models are used to generate key facility, safeguards, and security metrics along with SSBD recommendations. They are informed through several higher-fidelity capabilities including experimental testing, performance of measurement technologies, more detailed unit and measurement models, statistical methodologies, etc.

Flowsheet modeling is used to design the overall facility and defines flowrates, inventories, separation efficiencies, and batch timing. The flowsheet design takes into account both safeguards and security aspects in an iterative manner.

Once the flowsheet is defined, safeguards modeling is used to set up materials accountancy systems, process monitoring, and the various analytical methods to calculate material balances and set alarm conditions for material loss or misuse. Key safeguards metrics that are generated include estimates of Material Unaccounted For (MUF), the overall error ( $\square_{MUF}$ ), and detection probabilities for various diversion or misuse scenarios.

Finally, security modeling is used to lay out the physical protection system in 3-D and allows for force-onforce combat simulation. Three-dimensional modeling is required to capture the timelines for both outsider and insider scenarios. Facility layout is also a part of this work. Key security metrics include probability of adversary success against various theft or sabotage scenarios.

The initial demonstration of these capabilities for SSBD uses a generic electrochemical processing facility for processing of spent light water reactor fuel as an example. This example was chosen partly because the MPACT program has supported more research in electrochemical safeguards recently, and because commercial-scale facilities do not exist yet. Any potential future use of electrochemical reprocessing of spent fuel will benefit from SSBD recommendations now as opposed to farther along in a facility design process.

The capabilities shown on the left of Figure 23 make up the bulk of the research and development and provide high-quality data to the systems-level models. The MPACT program has a long history of funding new measurement technologies for materials accountancy applications. Several measurement technologies have been developed or tested for Electrochemical processing including the triple bubbler, voltammetry, microfluidic sampler, microcalorimetry, high dose neutron detector, and electrochemical sensor. Measurement models are used to determine how well the technologies will perform in production-scale facilities. Various statistical and detailed unit operation models provide more modeling detail for the systems-level models. Finally, radiation signatures and consequence models are used for facility and physical protection system design.

The capabilities are linked and used iteratively. For example, limitations of measurement technologies may steer the flowsheet model into a form that is more easily safeguarded. Information about the flowsheet and radiation signatures can affect the physical protection system since some areas of the facility have a high degree of self-protection. The choice of the final safeguards design will depend on the performance of the measurement technologies. In this manner, all capabilities work together to develop optimal safeguards and security designs and provide SSBD recommendations for facilities designers.

Safeguards Modeling: The Separation and Safeguards Performance Model is one of the systems-level modeling capabilities used for the overall design of a materials accountancy strategy. However, the modeling results are only as good as the data fed into the model, so the analysis work is strongly dependent on modeling and experimental work in the rest of the MPACT program, which provides more detail on the facility, measurement technologies, and overall safeguards approaches. A key aspect of this work has been the integration and coordination of data with other laboratories.

In 2019, the model was updated to be consistent with a baseline flowsheet, developed by ANL. A few modifications were required to make this change, and there will be some minor implications on the safeguards analysis. In addition, simulation data was generated for LANL to assist in the advanced integration tasks. That work takes a deeper dive into the performance of key MPACT technologies given the materials and environments they will face in a commercial electrochemical facility. In turn, the advanced integration work can then provide input to the Separation and Safeguards Performance Model such as expected measurement uncertainties.

In 2020, the final safeguards analysis will be completed based on the best information available about the measurement technology performance. The safeguards design will be presented along with its performance under diversion scenarios. The goal of this past year's work was to prepare the model for the final analyses.

**Security Modeling**: In 2019, a suite of tools, including Scribe3D and Blender, were used to model a generic Echem facility (Figure 24). This modeling work enables an analyst to optimize a security design for a new facility to avoid the high cost of retrofitting security elements to a facility after the design is complete. Physical protection elements such as sensors, portal monitors, barriers, and guard forces were added to the model based on best practices for physical security.

In order to understand the security performance of the system design, a vulnerability assessment was performed. Using a threat spectrum (4/5/6/7/8 adversaries), an outsider theft scenario was modeled to test the system. Utilizing a containment strategy, the response was able to keep material from being stolen from this hypothetical Echem facility design 75% of the time or greater for threats of six (6) or lower (see Figure 24). This was while only maintaining a security staff of 10 responders on site, and assuming a single offsite local law enforcement agency team of two (2) responders in 10 minutes. General best practice is to maintain a 3-to-1 ratio between responders and design basis threat in order to secure system effectiveness. Results for threat levels higher than six (6) were 68% effectiveness at seven (7) adversaries and 31% effectiveness at eight (8) adversaries, revealing that the system fails gradually, rather than suffering a steep drop-off at any single step. This is a useful data point when considering the possibility of attacks that may exceed the design basis threat. The system, as designed, offers some protection against large-scale threats.

The analysis provided some preliminary observations which are key aspects of Security by Design. A traditional double-fence Perimeter Intrusion Detection and Assessment System was not needed and would not have helped the responders. The thick shielding walls in the facility provide additional barriers to acquiring material and made it difficult for adversaries to breach the facility. Containment was found to be an optimal strategy as opposed to engaging adversaries when they are covering stairwells. In 2020, these observations will be expanded.



Figure 24. Echem facility 2/3D images.

#### Electrochemical Process Monitoring for Enhanced Safeguards:

In 2019, the MPACT campaign continued development of several process monitoring tools for application to Echem including a bubbler, an OR voltammetry probe, an electrorefiner (ER) voltammetry probe, an actinide sensor, and a U/TRU Pu measurement system.

Echem Process Monitoring – Bubbler: The ER step of the Echem process represents one of the biggest safeguard challenges. The ER contains large amounts of Pu in molten salt, making quantitative measurements exceedingly difficult. One proposed approach is to collect analytical chemistry samples and calculate the total Pu content given the ER salt density and volume. The MPACT research team has developed a bubbler system that can measure the ER density and height, enabling total Pu-mass calculation for a calibrated ER vessel.

In 2019, the bubbler that had been clogged in 2018 was cleaned and then re-installed into the JFCS IRT ER. Prior to insertion, a modified height gauge was installed in the HFEF to measure the physical characteristics of the bubbler as well as the ER vessel depth (Figure 25). After the bubbler was installed, it was continuously monitored for approximately 1000 hours. During that time, independent depth samples were taken and compared to bubbler determined salt depths. The percent difference between the measurements was 0.03% while the relative percent uncertainty on the bubbler measurement was 1.22%. The relative percent uncertainty for density and surface tension were 0.54% and 12.4%, respectively. In addition, key systematic errors were identified with the differential pressure transducers, including line losses at different flowrates.

During the above testing, two focused tests were conducted. First, the depth of the ER was varied using a dipper approach. Next, the salt temperature of the ER was varied. In the first test, a total of 821 g of salt was removed while monitoring the process via the triple bubbler. After the salt was removed, an

independent depth measurement was taken and compared to the bubbler value. The percent difference was under 1%. The salt was then added back into the ER while monitoring the process using the triple bubbler. In the salt temperature test, the salt density and surface tension shifted. No independent measurements were taken, but the bubbler appeared to track the changes appropriately.

Based on the testing, the systematic and random uncertainties of the triple bubbler and supporting instrumentation were evaluated. The largest source of error originated from the differential pressure transducers. Overall, 50% of the error came from a result of systematic uncertainties. Results from the uncertainty analysis as well as the triple bubbler testing met the 2019 Level 3 Milestone "M3FT-19IN04104042" titled "Estimate Accuracy and Precision."

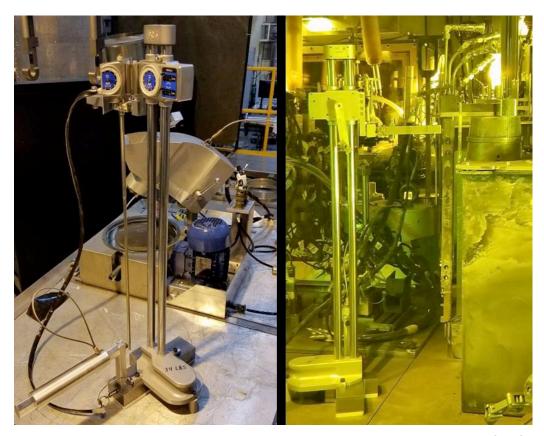


Figure 25. Triple bubbler salt depth height gauge with resistivity contact sensor, (left) in mockup and (right) in the HFEF hot cell.

**Echem Process Monitoring** – **Oxide Reduction Voltammetry**: During the Echem OR process, U and Pu should not migrate from the fuel basket into the OR salt in any appreciable amount. Monitoring that the OR salt is free from U and Pu is a strong indicator that the process is being run as declared, and valuable to the overall safeguards system. The MPACT research team has developed an OR voltammetry probe to demonstrate this capability and explore viability for long-term operation in the corrosive OR environment.

In 2019, the OR voltammetry sensor developed and tested in 2018 was further tested in the IRT OR located in INL's HFEF (Figure 26). The tests conducted were not consistent with the previous

laboratory experiments, nor with independent cyclic voltammetry (CV) scans obtained in the salt. Consequently, an electrical short is suspected to exist between one of the electrodes in either the sensor, the safeguards junction box, or the electrode cabling.

set of laboratory experiments was also conducted this year to refine the OR sensor design and answer questions about current electrode material selections. The main objectives of the experiments were to: (1) explore replacement options for the fragile and slow responding nickel/nickel oxide (Ni/NiO) reference electrodes, (2) explore replacement electrode materials for the tungsten (W) electrode that reacted in the IRT OR salt, and (3) characterize iridium (Ir) response to varying concentrations of lithium oxide (Li2O) (previously only studied at 1 wt%).

Bare Ir and Ni wires were selected as possible replacements for the Ni/NiO RE. These materials would function as pseudo-RE, which are thought to be less stable; however, testing demonstrated that both the Ir and Ni RE had less than 40 mV of shift over time, which is sufficiently stable. In addition, these materials are less finicky and respond rapidly to salt concentration changes.

Working electrode (WE) materials explored in the laboratory testing were molybdenum (Mo), 316 stainless steel (SS), and Ir. Short-term testing of the Mo WE indicated that the material would perform well in the OR salt. However, after constant immersion for several days, metallic dendrites, thought to be iron (Fe), formed on the Mo surface. Clear pitting of the Mo occurred as a result. As there are ppm levels of iron trichloride (FeCl3) in the OR, it is probable that formations like this would also occur if deployed as part of the sensor. The SS WE did provide decent CV measurements and was useful when comparing CV with independent measurements. In addition, it did not show significant signs of pitting or degradation. During testing of the Ir WE, two different coatings on the surface of the metal were observed. The first was a dark coating that is likely iridium oxide (IrO2). This coating was found to form only when operating the WE at the Ir oxidation potential in the presence of Li2O. The second coating formed chemically over time and was a dull white color. Means to remove the coating were not identified; however, the coating appeared to be stable and did not significantly affect the CV results.

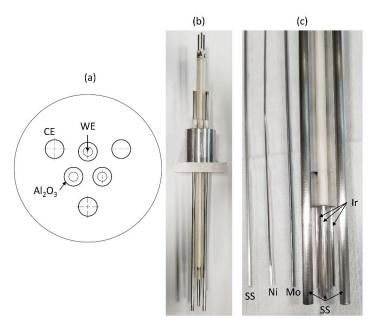


Figure 26. OR voltammetry probe installed in the IRT in HFEF.

Based on the field and laboratory testing, a second engineering-scale OR sensor was constructed and is ready for testing in the IRT ER. Key changes to the sensor over the previous one are: (1) one of the two Ni/NiO RE has been replaced with a bare Ni wire, (2) the porous plug thickness in the Ni/NiO RE has been reduced from 25 mm to 12 mm (should make it more responsive), (3) the W WE was replaced with a SS WE, and (4) modifications were made to the housing to ensure the sensor is isolated from the OR vessel. These modifications will be tested in 2020.

*Echem Process Monitoring – Electrorefiner Voltammetry*: For the reasons mentioned above, the ER presents significant safeguards challenges. In addition to analytical chemistry-based accountancy techniques, MPACT has pursued ER voltammetry to provide real-time qualitative, and potentially quantitative, information about the ER salt composition.

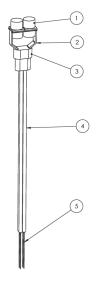
The objective of ANL's 2019 research was to integrate a multifunctional voltammetry sensor capable of measuring actinide concentrations and the salt potential into the operations of an engineering-scale electrorefiner. The measurements provided by the sensor are critical for material accountancy and process monitoring for molten salt reprocessing equipment. The work was successfully accomplished, and the sensor was able to provide high-quality measurements even as high current processing operations were simultaneously conducted by the electrorefiner. Use of the sensor was optimized to provide the best balance of safeguards and process control measurements while ensuring that operations of the electrorefining process could proceed unimpeded. The totality of data from long-duration testing in 2019 and 2018 were analyzed to provide an assessment of the stability, accuracy, uncertainty, and lifetime that can be expected from electroanalytical sensors used in large-scale electrorefining equipment. Finally, preparations were made to supply a voltammetry sensor to INL in order to enable monitoring capabilities for the IRT electrorefiner that is operating in the hot cell environment at the HFEF. A version of the multielectrode sensor was delivered to INL in mid-September and is awaiting qualification within the mock-up area.

Much of the work in 2019 has involved preparations for use of the voltammetry sensor in INL's IRT JFCS electrorefiner. In order to demonstrate safeguards capabilities in the IRT ER, a number of modifications to the sensor needed to be made to address complications that may arise from use in the hot cell environment.

The differences between the ANL in-house sensor and the sensor developed for INL involve geometric differences in the instrumentation ports, wiring, and general hot cell encumbrances. The sensor is to be installed into one of the instrumentation oblong ports (obports). These obports are only 5/8" in width, which necessitated a reduction in size of the ANL sensor platform. This requirement stands in contrast to the need for greater mechanical robustness from the sensor in order to ensure that it does not break given the rough handling that happens in hot cells where the sensor must be maneuvered by manipulators. In order to satisfy these contrasting demands, the use of a shroud had to be eliminated from the sensor design in order to permit the use of thicker electrode support materials. The absence of a shroud will make the sensor more sensitive to convection effects, but it is believed that these issues will be manageable.

A final design sketch of the sensor developed for INL is shown in Figure 27. This new design fits into the obports and is completely electrically isolated from the electrorefiner's upper assembly. The sensor can be fully disassembled such that each of the individual electrodes can be removed and replaced if necessary. Amphenol connectors now provide electrical connections to the tungsten electrodes, as opposed to alligator clips as used in ANL's earlier designs. These connectors should facilitate easier connections and

disconnections of the wiring when using manipulators. This modified probe was manufactured at the end of 2019, sent to INL for qualification, and will be tested in HFEF starting in 2020 Q2/Q3.



- 1. Amphenol-type circular connector
- 2. Strain-relief wiring bracket
- 3. Sensor body (stainless steel)
- 4. Alumina tubes (0.25" diameter)
- 5. Tungsten electrodes

Figure 27. Schematic diagram of voltammetry sensor for use in the IRT electrorefiner.

Echem Process Monitoring – Actinide Sensor: In pyroprocessing of spent nuclear fuels, uranium metal and other active transuranics are oxidized and dissolved from an anode into a LiCl-KCl-UCl3 salt electrolyte. Uranium is simultaneously reduced from the electrolyte to a cathode and collected as high purity uranium, which is referred to as an ER process. Transuranics accumulate during the ER process over time. Because the conventional chemical analysis of ER salt by inductively-coupled plasma mass spectrometry is inconvenient and usually time-consuming, it is desired to monitor the actinides in the salt in real time for safeguard purposes.

The actinide sensor under development is based on the potentiometry of an electrochemical cell formed between a U- $\beta$ " alumina membrane tube and an Ag/AgCl reference electrode in LiCl-KCl-UCl3 salt. The U- $\beta$ " alumina membrane is made by ion exchange of a Na- $\beta$ " alumina tube (procured from Ionotec in UK) in UCl3 salt and is U3+ ion selective, in theory. The activities in 2019 were mainly focused on a LiCl-KCl-UCl3 salt system, including development and characterization of U- $\beta$ " alumina membrane made by ion exchange of Na- $\beta$ " alumina in UCl3 salt, selectivity and repeatability of U- $\beta$ " alumina for UCl3 monitoring in LiCl-KCl-UCl3 salt, and design of an engineering-scale U-sensor for ER salt in the JFCS project.

A systematic study was performed on U-  $\beta$ " alumina membranes fabrication by ion exchange of Na-  $\beta$ " alumina in solid UCl3 powder at different temperatures (i.e. 650°C, 680°C, and 735°C) and in UCl3 vapor at 920°C with Na- $\beta$ " alumina tube sample above UCl3 liquid. Results showed that (1) when the ion exchange temperature was 650°C and 680°C, high stresses accumulated from the ion exchange process could lead to shattering or cracking of U-  $\beta$ " alumina during the water cleaning stage; and (2) when the ion exchange temperature was 735°C, no visible hairline cracks were observed, and the chemical reaction between Na-  $\beta$ " alumina and UCl3 was slight. Although ion exchange at 735°C could lead to a decrease of strength of the U- $\beta$ " alumina due to stress accumulation, the subsequent annealing process at 800°C could

help relieve the stress and increase the strength. Extended ion exchange time could increase the chemical reaction and weaken the fabricated U- β" alumina membrane. The ion exchange at 920°C UCl3 vapor was very successful—no chemical reaction between Na-β" alumina and UCl3 vapor was observed, the mechanical strength of the U-β" alumina after ion exchange was significantly higher than that made by ion exchange in 735°C solid UCl3 powder. No water cleaning is needed, and the UCl3 salt used for ion exchange could be recycled and reused (Figure 28).



Figure 28. Picture of components for assembling a U-sensor electrode and a U-sensor electrode ready for testing.

Because a high-quality, crack-free U- $\beta$ " alumina membrane is critical for reliable, repeatable monitoring of UCl3 salt, some efforts were spent on the ion exchange of high strength Na- $\beta$ " v alumina samples—with a fracture strength of ~900MPa and procured from Materials & Systems Research Inc., in parallel with the U-sensor development work using the Ionotec Na- $\beta$ " alumina materials. The ion exchange of the high strength Na- $\beta$ " alumina in UCl3 salt was successful and it also exhibited good compatibility in LiCl-UCl3 salt, greatly contrasting to the Ionotec Na- $\beta$ " alumina, which exhibited poor compatibility in LiCl-KCl-UCl3 salt.

U-sensor testing results showed that the U- $\beta$ " alumina can respond to the UCl3 concentration changes and its selectivity is reasonably good. When the system reached the needed equilibrium state, the measured open circuit potential difference due to UCl3 concentration changes was consistent with the Nernst equation, but the repeatability could be improved. It is expected that the improvement in the mechanical strength (or minimization of strength loss) and microstructure of U- $\beta$ " alumina (no microcracks) immediately after ion exchange could help improve the performance of the U-sensor for ER salt.

**Echem - U/TRU Pu Measurement**: The U/TRU metallic ingot product from the Echem process is of particular safeguards concern. At commercial scales, this product will nominally contain kgs of 50% Pu and 50% U, and has had most of the fission products removed, significantly raising the material attractiveness. Additionally, traditional nondestructive assay techniques are unproven and have several known deficiencies for this application. Finally, analytical chemistry sampling may be limited by

homogeneity issues on the solid product. One proposed method for measuring plutonium content in U/TRU metallic ingots utilizes the unique thermophysical properties (i.e. the solidification curve) of the U/TRU alloys. Thermocouple measurements can be taken during solidification of the U/TRU ingots by monitoring the temperature of the melt and/or crucible. An inflection is indicated on the time versus temperature solidification curves caused by the latent energy evolved from the alloy on freezing relative to normal heat transport conditions. Assuming a U-Pu alloy, the inflection point is directly related to the plutonium concentration in the U-Pu alloy. The temperature at the inflection point can then be translated to liquidus data from established U-Pu phase diagrams for the determination of Pu content. In 2019, three U/TRU ingots produced during the JFCS IRT were utilized for Pu measurements using this method.

For these tests, specific quantities of pure aluminum and silver were utilized for the calibration of internal thermocouples in the range of heat characteristics expected for testing with the U-TRU alloys. A small furnace applicable to U/TRU masses in the 100 g range was installed in HFEF and proved capable of measuring the liquidus or melting point of three U-TRU alloys from which a plutonium content of the alloy was determined (Figure 29). For the three alloys tested, there appears to be a slight bias in the calculated-to-measured % Pu to overpredict the Pu content, even outside the analytical error of + 5%. The oxidation of metallic pieces during sampling/analyses may be the cause of the lower than expected chemistry results. The lower mass or numerous heats on the first U-Pu alloy may have contributed to the large difference in calculated-to-measured data. The success of this technique resulted in its transition to the JFCS, and its experimental results will be utilized as part of the safeguards evaluation in both the JFCS and MPACT Milestone 2020.

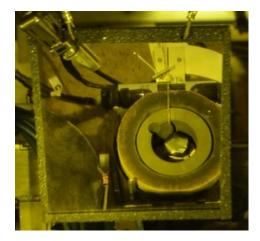


Figure 29. U/TRU measurement system installed in the IRT in HFEF.

#### 6.2.2 Exploratory Research/Field Tests

The MPACT research team continued to develop instruments with new capabilities that will advance the state of the art in nuclear material accounting and control. A focused, innovative, engineering-driven science-based R&D program was conducted in 2019 to improve precision, accuracy, speed, sampling and monitoring methods, and to extend the usable operational range of two MPACT-developed nuclear material accounting and control technologies. As the technical readiness level of these technologies increased, the team's focus shifted toward executing field tests in fuel cycle facilities to obtain operational experience and demonstrate effectiveness.

*Microcalorimetry*: The MPACT research team's development of the super-high-resolution gamma spectrometer based on microcalorimetry concluded in 2018, and MPACT personnel focused on preparing the instrument for field trials in 2019. Samples from across the U.S. National Laboratory complex were collected and measured at LANL as part of a user assessment. The goal of this effort was to (1) understand the full range of safeguards information that could be extracted from microcalorimetry measurements of samples taken from as many U.S. nuclear fuel cycle processes as possible, and (2) assess the operation of the LANL microcalorimetry from an end-user perspective. In 2019, Echem samples were unavailable, but they are expected in 2020 and will be analyzed when received at LANL.

Our experience with the 2019 User Assessment and Measurement Campaign demonstrated that the technology readiness level of microcalorimeter gamma spectroscopy has significantly advanced over the past several years. Advances in both instrumentation and data analysis are now making acquisition of high-quality spectra routine, enabling new safeguard capabilities. The measurements shown here represent the largest set of ultra-high-resolution, high-statistics microcalorimeter gamma spectra ever recorded. The spectra are now being used to explore signatures of spent fuel, explore uncertainty limits of nondestructive uranium and plutonium isotopic characterization, inform safeguards models, and extract improved nuclear data.

With upgrades to the adiabatic demagnetization refrigerator control system and data acquisition software, Source Finding Application (SOFIA) will be ready for deployed measurement campaigns. Being able to bring a microcalorimeter gamma spectrometer to specific measurement locations (for example, HFEF at INL or ORNL spent fuel hot cells) opens up important opportunities to acquire data needed to inform safeguard models and answer questions about nuclear facility applications.

The SOFIA software also demonstrates the maturity of the high-throughput microwave multiplexing architecture. In 2020, we are planning to build a new gamma spectrometer with the same architecture for use in the INL analytical lab. With a modular design of 300–500 pixels and a larger dilution refrigerator capable of continuous operation below 100 mK (no regeneration needed), this spectrometer will maximize performance for dilute samples that require long counting times.

As practical microcalorimeter spectrometers are implemented at nuclear facilities and analytical laboratories, their rapid and nondestructive measurement capabilities are expected to reduce reliance on sampling, mass spectrometry, and destructive chemical analysis. As a result, safeguards and security goals can be achieved with reduced cost and impact on facility operations. We anticipate that microcalorimeter technology will be an important component of an advanced safeguards approach to enable the next generation of nuclear energy.

In 2020, microcalorimetry efforts will be focused on deployment at INL facilities. Additionally, 2020 microcalorimetry activities will focus on IAEA Siebersdorf Analytical Lab applications. These efforts will be funded by the U.S. support program to the IAEA, and developed out of the data collected by the MPACT campaign (Figure 30).









Figure 30. (Left to right) Photos of ATM-109-5, ATM-109-10, Sister Rod sample, and Z10924 measured during the 2019 measurement campaign.

*High Dose Neutron Detector*: Nondestructive assay of nuclear material relies heavily on neutron detection to assay for U and Pu content. The ability to accurately measure correlated neutrons in the presence of high gamma fields will be critical to the successful assay of U and Pu in advanced fuel cycles where fission products remain at some level with the special nuclear material. The MPACT research team developed a High Dose Neutron Detector (HDND) to address these issues.

For 2019, the MPACT HDND (Figure 31) activities transitioned from development to field testing, including a performance evaluation for high mass Pu items representative of Echem U/TRU product ingots. The measurement campaign involved a range of Pu-U-Mo (PUMH) plates in configurations with increasing number of PUMH plates to evaluate HDND performance for range of Pu-mass items (Figure 32). The maximum number of plates (10) corresponded to nearly 2.3 kg of Pu, which provided an opportunity to characterize HDND for materials with Pu-mass in the range expected for commercial-scale U/TRU ingot.

The measurement campaign demonstrated HDND capability to perform coincidence (doubles) and even multiplicity (triples) counting in a two-HDND-unit configuration. This represents a remarkable result, given only part of the detection cells (approximately one-half from the two units) was functional. Note that more than half the detection capability (several cells in two HDND units and most cells in the third unit) was lost due to a power surge during the preparatory stage of the campaign. A system of three fully functional HDND units would, therefore, provide a feasible multiplicity counting capability. Furthermore, the measurements demonstrated capability of HDND to measure multiplication (and hence Pu content) for high mass Pu-bearing items through *D/S* ratio as well as capability to perform singles counting on individual detection cells, which provide access to back-to-front ratio. The use of back-to-front ratio provides an attractive technique to assay Pu content of U/TRU ingot as it does not require the high-detection efficiency necessary for correlated (i.e. doubles) counting. The back-to-front ratio is expected to provide access to Pu content in a single HDND pod (or miniHDND) configuration.

In 2020, this technology has been transferred to the JFCS for inclusion in the safeguards evaluation.



Figure 31. Two HDND panels set up with PUMH plates used in Zero Power Physics Reactor (ZPPR) measurement campaign.

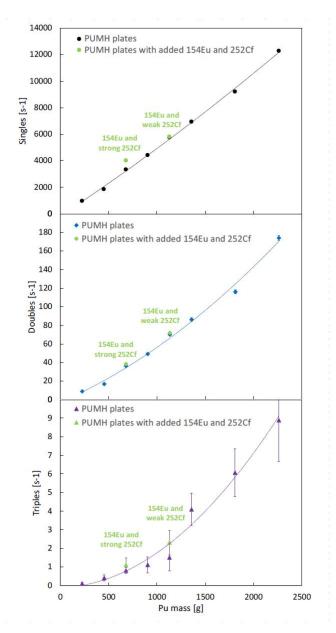


Figure 32. Two HDND pod measured singles, doubles, and triples count rates as a function of Pu-mass for series of 1–10 PUMH plates (note, measurements with 7 and 9 plates were not performed).

Versatile Test Reactor Program

### 7. Versatile Test Reactor (VTR)



#### **Director's Message**



The Versatile Test Reactor (VTR) Program has experienced tremendous change, progress, and growth throughout 2019, notably moving toward a DOE Order 413.3B capital acquisition project after the approval of the mission need. Highlights of major accomplishments spanning through September 2019 are included in this report. The near-term priorities include preparing the Conceptual Design Report and the Conceptual Safety Design Report, preparing the cost estimate and schedule, and achieving approval of Critical Decision (CD)-1, *Approve Alternative Selection and Cost Range* in 2020. A strong national team is established, including multiple national laboratories, industry partners, and universities.

## Critical Decision-0 Approval

Energy Systems Acquisition Advisory Board members unanimously voted to approve CD-0, *Approve Mission Need*, on February 22, 2019, which was subsequently approved by the deputy secretary of energy. Secretary Perry announced the launch of the Department of Energy's Versatile Fast Neutron Source, also referred to as the Versatile Test Reactor, one of the foundational projects specified in the Nuclear Energy Innovation Capabilities Act of 2017.

### VTR Multi-Laboratory Memorandum of Understanding

The VTR multi-laboratory Memorandum of Understanding (MOU) was signed between INL and ANL, LANL, ORNL, Pacific Northwest National Laboratory, and Savannah River National Laboratory. This MOU indicates full partnership for each laboratory and establishes a mutual understanding for accomplishing the technical work scope and



The Deputy Secretary of Energy

February 22, 2019

MEMORANDUM FOR MARK W. MENEZES UNDER SECRETARY OF ENERGY

FROM: DAN BROUILLETTE

SUBJECT: Approval of Critical Decision-0, Approve Mission Need, for the Versatile Test Reactor (VTR) Project

As Chief Executive of Project Management, I approve Critical Decision (CD)-0, Approve Mission Need, for the Versatile Test Reactor (VTR) project with a cost range of \$3.0B to \$6.0B and an expected CD-4, Project Completion, range of 2026 to 2030.

The VTR project will provide leading edge capability for accelerated testing and qualification of advanced fuels and materials, enabling the United States to regain and sustain technology leadership in the area of current and future advanced reactor systems

At the Energy Systems Acquisition Advisory Board (ESAAB) meeting on February 21, 2019, the members strongly supported this project and unanimously voted to approve CD-0 for the VTR project. With their endorsement, I approve CD-0.

ec: E. McGinnis, NE-2 D. Miotla, NE-20

D. Miotla, NE-20 J. Herczeg, NE-4 B. Singh, NE-42 M. Peek, PM-1

R. Hendrickson, CF-2

Figure 33. Critical Decision-0 Approval

the cost and schedule baseline, as well as committing resources as necessary to execute approved work plans.

#### **Conceptual Design Subcontract**

A subcontract was issued to GE-Hitachi/Bechtel in November 2018 to support the pre-conceptual and conceptual design efforts for the VTR. The conceptual design is based on GE-Hitachi's power reactor innovative small module (PRISM) Mod-A reactor design, with necessary modifications to meet VTR needs.

Other industry contracts are also in place to support specific activities in the VTR design and engineering. A reference core design was completed by the national laboratories to support the overall conceptual design effort.

#### **Engineering Model**

The engineering model has been successfully moved to Aveva, a digital engineering software suite. The suite supports engineering analysis, a physical model, construction sequencing, and quantities for cost estimating. It is the culmination of software selection and deployment, and placement of thousands of inputs to the model.

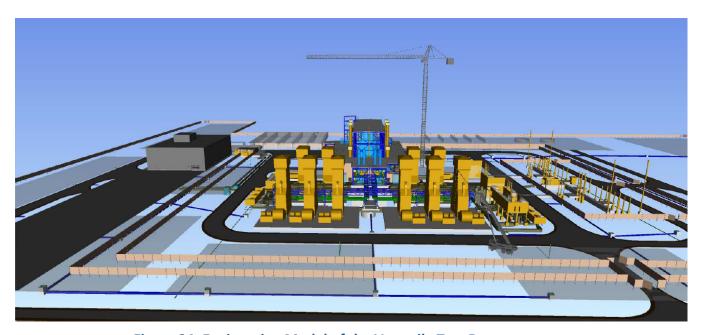


Figure 34. Engineering Model of the Versatile Test Reactor.

#### Safety Design Strategy Approval

Final approval of the safety design strategy was received from the Department of Energy-Idaho Operations Office on July 1, 2019. The safety design strategy provides the general approach and requirements that will be used to ensure VTR is designed and operated safely. It provides the first basis for outlining how VTR will use a risk-informed approach to the development of the VTR safety case and safety basis documents.

The safety design strategy is a required deliverable as part of the CD-1 package and was completed nearly one year ahead of the planned CD-1 submittal.

#### **Environmental Impact Statement (EIS)**

The Notice of Intent to prepare an environmental impact statement for the VTR was published in the Federal Register on August 5, 2019, with a public comment period continuing through September 4, 2019. Web-based public scoping meetings were held August 27 and 28, 2019. Four videos were produced and presented during the scoping meetings to inform and educate the public. Multiple comments were received and are being addressed by DOE.

#### **MOU** with the NRC

The VTR will be designed, built, and operated under a DOE authorization. However, a MOU is signed with the NRC to participate in the VTR effort as an observer, in preparation for the NRC to license subsequent commercial fast reactors.

#### **International Collaborations**

An implementing agreement with France and a Memorandum of Collaboration with Japan have been signed to enable information exchange and collaborative activities specific to VTR.

#### **New University Partners**

Six university-led projects were awarded \$1.7 million in funding to support experiment vehicle development for the VTR. A total of 20 universities and nine industry partners now collaborate with the VTR in nine different areas: Sodium Reactor, Molten Salt Reactor,

Figure 35. Participants of the VTR workshop with Japan, September 10–11, 2019.

Structural Materials, Rabbit Systems, Lead/LBE Reactor, Data and Modeling and Simulation, Gas Fast Reactor, Virtual Design and Construction, and Strategic Initiatives. The following map depicts all of the VTR collaborating organizations.



Figure 36. VTR Partnership Map.

- Industry Partners: Bechtel, The Cameron Group (TCG), Electric Research Power Institute (EPRI), Framatome, General Atomics (GA), General Electric-Hitachi (GEH), The HDF Group (HDF), TerraPower, Westinghouse
- ★ University Partners: Abilene Christian University (ACU), Colorado School of Mines (MINES), Fort Lewis College (FLC), Georgia Tech (GT), Idaho State University (ISU), Illinois Institute of Technology (IIT), Massachusetts Institute of Technology (MIT), North Carolina State University (NCSU), Oregon State University (OSU), Purdue University, Texas A&M University, University of California, Berkeley, University of Idaho (U of I), University of Michigan (UMich), University of New Mexico (UNM), University of Pittsburgh (PITT), University of Utah (U of U), University of Wisconsin (UW), Virginia Commonwealth University (VCU), Yale University
- ▲ National Laboratories: Argonne National Laboratory (ANL), Idaho National Laboratory (INL), Los Alamos National Laboratory (LANL), Oak Ridge National Laboratory (ORNL), Pacific Northwest National Laboratory (PNNL), Savannah River National Laboratory (SRNL)
- Department of Energy (DOE)

# Appendix A Acronyms

AFC Advanced Fuels Campaign

ANL Argonne National Laboratory

APS Advanced Photon Source

ATF accident tolerant fuel

ATR Advanced Test Reactor

ART Advanced Reactor Technologies (program)

BEV battery electric vehicle

CD Critical Decision

CEA French Alternative Energies and Atomic Energy Commission

CoDCon Co-Decontamination Project

DDII Development, Deployment, and Implementation Issues

DOE Department of Energy

DOE-NE DOE Office of Nuclear Energy

ER electrorefiner

ESP Equilibrium System Performance

EURATOM European Atomic Energy Community FCF Fuel Conditioning Facility

FCRD Fuel Cycle Research and Development

GCAM Global Change Assessment Model

GE General Electric

HALEU High-Assay Low-Enriched Uranium

HDND High Dose Neutron Detector

HFEF Hot Fuel Examination Facility

HFIR High-Flux Isotope Reactor

IAEA International Atomic Energy Agency

INERI International Nuclear Energy Research Initiative

INL Idaho National Laboratory

IRT Integrated Recycling Test

JAEA Japan Atomic Energy Agency

JFCS Joint Fuel Cycle Studies

KAERI Korean Atomic Energy Research Institute

LANL Los Alamos National Laboratory

LTA lead test assembly

LWR light water reactor

MC&A material control and accounting

MPACT Materials Protection, Accounting, and Control Technologies

MRPP Material Recovery Pilot Plant

MRWFD Material Recovery and Waste Form Development

MUF Material Unaccounted For

NE-4 DOE Office of Nuclear Technology Research and Development

NEA Nuclear Energy Association

NEAMS Nuclear Energy Advanced Modeling and Simulation

NES nuclear energy systems

NFCSC Nuclear Fuel Cycle and Supply Chain

NNSA National Nuclear Security Agency

NRC Nuclear Regulatory Commission

NSSS Nuclear Steam Supply System

NTD National Technical Director

OECD-NEA Organization for Economic Cooperation and Development-Nuclear Energy Agency

OR Oxide Reduction

OR-SAGE Oak Ridge Siting Analysis for power Generation Expansion

ORACLE Oak Ridge Advanced Fuel Cycle Logistics Environment

ORNL Oak Ridge National Laboratory

PNNL Pacific Northwest National Laboratory

PUMH Pu-U-Mo

PWR pressurized water reactor

R&D research and development

RD&D research, development, and demonstration

SA&I Systems Analysis and Integration Campaign

SNS Spallation Neutron Source

SOFIA Source Finding Application

SSBD Safeguards and Security by Design

SSC System Steering Committee

SSEC selection and evaluation criteria

TREAT Transient Reactor Test facility

TSRA Technology and System Readiness Assessment

U.S. United States

U/TRU uranium with transuranic waste

UNF used nuclear fuel

VTR Versatile Test Reactor